Radiation stability of ¹²⁵I-brachytherapy sources

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Abstract. Low energy ¹²⁵I-brachytherapy sources are used for the treatment of retinoblastoma and many other forms of eye cancer. Such sources were prepared by adsorption of ¹²⁵I on palladium coated silver rods and were critically evaluated for safety aspects, as per AERB standards. In order to attain low leachability and to facilitate leak free laser encapsulation of sources within titanium capsules of size 0.8 mm (ϕ) × 4.75 mm (l), the radioactive source core was coated with polystyrene. With a view to study the radiation stability of such sources over a period of three weeks, both polymer coated radioactive sources and inactive source cores were separately subjected to an integrated gamma dose of ~(17.85 × 10⁴) Gy (17.85 MRad), which is the dose expected to be received in three weeks from a source containing ~ 111 MBq of ¹²⁵I. This was carried out to test their suitability for reuse within such period. SEM pictures of inactive source cores were evaluated and found to be satisfactory. The sources were found to be reusable safely, for repeated brachytherapy procedures over a practically useful time of at least three weeks.

Key words: ¹²⁵I-brachytherapy sources • retinoblastoma • radiation stability • SEM • leach rate

Introduction

¹²⁵I-brachytherapy sources in various shapes and sizes find extensive use in eye and prostate brachytherapy [2, 3]. Palladium coated silver rods of size 0.5 mm (ϕ) × 3 mm (l), containing ~(74–111) MBq of ¹²⁵I have already been developed and are being used in India as brachytherapy sources, for the treatment of various forms of eye cancers (Fig. 1). These sources in the low activity range of ~(18.5–37) MBq can also be used as permanent implants for the treatment of localized prostate cancer. Prior to their clinical application, the bare radioactive sources are sealed within 50 µ thick tiny titanium capsules using a Nd:YAG laser. To reduce the leachability of bare radioactive source and to make the outer surface of the source free from loosely

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I-125 adsorbed on palladium coated silver rod 0.8 mm 0.8 mm 4.75 mm held activity, the bare radioactive sources are coated with a thin film of polystyrene. The coating with polystyrene does not significantly reduce the dose output from low energy sources of 125 I, emitting ~(27.4–35) keV X-rays/gamma rays and renders the source safe for handling. During the period of clinical use, the sources used for the treatment of one patient may again be used for the treatment of another patient, provided the source activity is adequate for another treatment. In case of eye brachytherapy, although the sources are used in a localized region and the radioactivity level involved with each source is not very high, there is still a probability that radiation damage of the polymer coating due to the decay heat of ¹²⁵I may lead to increase in leachability and also may result in non-uniformity of ¹²⁵I within the source core. Hence, the radiation stability of sources due to the decay heat needs to be studied over a period of their practically useful life of about three weeks. In the present communication, we describe our studies that were carried out to study the effect of gamma radiation on palladium coating of inactive source cores and also on the thin polystyrene coatings applied over the outer surface of radioactive ¹²⁵I-brachytherapy sources. The results obtained were found to be useful for evaluation of the practical utility of sources for repeated brachytherapy procedures.

Materials

Reducing agent free ¹²⁵I as sodium iodide in dilute NaOH solution was procured from Radiochemicals Section of our Division. High purity silver wire of $\sim 0.5 \text{ mm} (\phi)$ was procured from M/s Silver Plaza, Mumbai. Polystyrene beads of $\sim 6 \text{ mm}$ (ϕ) were procured from M/s Fluka Chemicals, Switzerland. A well type re-entrant ion chamber used for measurement of radioactivity was procured from M/s SUN Nuclear Corporation, USA. SEM micrography of inactive samples was carried out with a JEOL, JSM-T 330 scanning electron microscope in the Chemistry Division of our Institute. The samples were irradiated in a GC-5000 gamma chamber at the Radiation Technology Development Section of our Institute. Optical density measurements of exposed X-ray films were made with the help of an OPTEL-B&W transmission densitometer. All other chemicals used in our work were of GR/AR grade procured from reputed manufacturers.

Experimental

The adsorption of ¹²⁵I on palladium coated silver has been earlier reported by us and is being used as an established method for the fabrication of ¹²⁵I-brachytherapy sources required for the treatment of eye cancer [7]. The sources were prepared by previously standardized protocol and polystyrene coating was done on bare radioactive sources. The sources were assayed and their leachability was determined. Subsequently, the radioactive sources were irradiated in a gamma chamber with an integrated gamma dose of ~(17.85 × 10⁴) Gy (17.85 MRad) and post-irradiation leachability and uniformity of activity was evaluated. Irradiation of few inactive palladium coated silver rods was also carried out with a similar dose and SEM micrographs of irradiated and unirradiated samples were compared to observe the visual changes on outer surface of inactive samples.

Preparation of ¹²⁵I-brachytherapy sources

Silver rod of $\sim 0.5 \text{ mm}(\phi)$ having purity greater than ~ 99% was cut into 0.5 mm (ϕ) × 3 mm (1) pieces and the cut pieces were cleaned with 3 M HCl followed by thorough wash with distilled water. The coating of palladium on silver rods was carried out by treating them with 0.05% PdCl₂ solution at ~100°C for 15–20 min. The coated rods were individually taken in small glass reaction tubes and subsequently treated with $\sim 40 \ \mu L$ of ¹²⁵I solution containing \sim 130 MBq (3.5 mCi) of ¹²⁵I. About 5 µg of carrier iodide as KI was also added to each individual reaction tube to enhance adsorption. The total reaction volume was maintained as 50 µL and the reaction temperature was maintained at \sim (60–70)°C for 6-8 h [7]. The activity of radioactive sources after the adsorption was measured by using an SNC-1008 Model (Manufactured by SUN Nuclear Corporation, Melbourne, Florida) re-entrant ion chamber duly calibrated in the Accredited Dosimetry Calibration Laboratory (ADCL), Department of Medical Physics, University of Wisconsin-Madison.

Polymer coating on outer surface of radioactive sources

The bare radioactive sources containing \sim (74–111) MBq (2–3 mCi) of ¹²⁵I were coated with polystyrene. Polystyrene beads of ~6 mm (ϕ) were dissolved in benzene at different concentrations. In order to have a thin and firm coating of polystyrene a concentration of 175 mg per mL was found to be the most suitable. The radioactive sources were individually dipped in polystyrene solution for ~10 s, dried and washed with luke warm water at ~(30–35)°C [6].

Gamma irradiation of ¹²⁵ I-sources

Even though the size of the source is very small and majority of X and gamma emissions are likely to pass through without depositing their entire energy within the source, it was presumed that all the decay heat resulting from ¹²⁵I-source is absorbed within the source itself. Total number of disintegrations from a typical ¹²⁵I-source of ~111 MBq (3 mCi) activity were calculated over a decay period of 60 days by using the following formula [5]:

(1)
$$N_T = N_0 \left(1 - e^{-\lambda t}\right) / \lambda$$

where: N_0 – initial number of disintegrations per unit time at time = 0; N_T – total number of disintegrations in time 't'; λ – disintegration constant; t – time elapsed.

Based on an earlier report, the average energy released per disintegration from the decay of ¹²⁵I, including the energy released by the low energy Auger electrons and conversion electrons, was assumed to be 61.44 keV [4]. Over a period of three weeks, total integrated absorbed dose due to the radioactive decay of ¹²⁵I-source, weighing ~ 10 mg and having the activity of ~111 MBq, was approximately estimated to be $\sim (17.85 \times 10^4)$ Gy (17.85 MRad). Few inactive palladium coated silver rods along with the radioactive sources of ~71.4 MBq (1.93 mCi) average activity were irradiated in the GC-5000 gamma chamber for a period of ~51 h. An integrated absorbed gamma dose of ~ (17.85×10^4) Gy (17.85 MRad) was imparted to all the samples at a rate of \sim 3500 Gy/h (0.35 MRad/h) by means of a high intensity radiation source comprising of ~129.5 TBq (3500 Ci) of ⁶⁰Co. The quantum of imparted gamma dose was confirmed by standard Fricke dosimetry.

SEM micrography of inactive samples

SEM micrographs of both unirradiated and irradiated inactive palladium coated silver rods were taken to observe the morphological changes due to gamma irradiation. The samples were cleaned and individually kept in a vacuum chamber of the instrument at a pressure of 5×10^{-6} torr. The electrons emitted by excitation of a tungsten cathode filament of the instrument (having an average energy of ~ 30 keV) were focused to fall on the samples up to a depth of $\sim 10 \ \mu m$ and secondary electrons emitted from the surface of samples, having energy in the range of 10–30 eV were utilized in a NaI(TI) detector coupled with a cathode ray tube to form an image. SEM micrographs of both irradiated and unirradiated samples were taken with 5000-fold magnification and compared for determining the changes in surface texture.

Leachability

The leachability of gamma irradiated and unirradiated ¹²⁵I-sources was determined under static conditions as per the AERB stipulated procedure [1]. Individual radioactive sources containing \sim 71.4 MBq (1.93 mCi) of ¹²⁵I were kept in 100 mL of distilled water for 48 h. The radioactivity released in water after the test was estimated by counting the samples of test solution using the well type NaI(Tl) scintillation counter set for ¹²⁵I measurement.

Uniformity of activity of gamma irradiated source

The uniformity of the activity of irradiated radioactive sources was assessed by a previously reported procedure [7]. After applying suitable decay corrections, individual radioactive source having \sim 39 MBq (1.05 mCi) of ¹²⁵I was kept in a glass tube and mounted on a specially fabricated autoradiography gadget. An industrial radiographic X-ray film was wrapped along the circumference of the gadget. The exposure time was optimized at ~ 20 min. Later, the exposed film was developed and optical density at eight different positions of the film was measured with the help of the B&W transmission optical densitometer.

Results and discussion

 125 I could be quantitatively (> 80%) adsorbed on palladium coated silver wires and sources containing ~74 MBq (2 mCi) of ¹²⁵I were coated with thin layers of polystyrene. Even though, the sources under study were having a radioactive activity of \sim 71.4 MBq (1.93 mCi) of ¹²⁵I, they were irradiated with higher quantum of integrated gamma dose that may be encountered in actual clinical practices while using a typical source incorporating maximum of ~111 MBq (3 mCi) of ¹²⁵I. Gamma irradiation of the inactive palladium coated silver rods and that of radioactive sources was carried out to impart an integrated absorbed dose of $\sim (17.85 \times 10^4)$ Gy (17.85 MRad). SEM micrographs of the inactive unirradiated and irradiated palladium coated silver rods under 5000-fold magnification indicated a mild agglomeration on the outer surface of irradiated samples, without affecting the firmness of palladium coating (Fig. 2). In a separate



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Fig. 2. SEM micrograph of unirradiated inactive Pd-Ag rod (a) and gamma irradiated inactive Pd-Ag rod (b).

| Source no. | Pre-irradiation leachability (%) | | Post-irradiation leachability (%) | | |
|------------|----------------------------------|------------|-----------------------------------|------------|--|
| | after 24 h | after 48 h | after 24 h | after 48 h | |
| 1 | 0.0023 | 0.003 | 0.0032 | 0.0039 | |
| 2 | 0.0026 | 0.0048 | 0.0028 | 0.005 | |
| 3 | 0.001 | 0.0021 | 0.0014 | 0.0023 | |

Table 1. Leachability of ¹²⁵I-sources exposed to 17.85×10^4 Gy dose produced by a ⁶⁰Co-source (source activity = 71.4 MBq (1.93 mCi) \pm 2.5%; temperature = 22–23°C (ambient))

study, when the sources were prepared with gamma irradiated palladium coated silver rods, this change was not found to adversely affect subsequent adsorption and uniformity of activity of ¹²⁵I. The details of such experiments are not included in the present text and shall be published elsewhere. The leachability of unirradiated and also of gamma irradiated radioactive sources was studied separately and in all the cases it was found to be well within the maximum limits (i.e., 0.01% of total activity of source) set by regulatory authority (i.e., AERB, India) with a maximum value of 0.005% (Table 1). This indicated that the integrity of the polystyrene coating and firmness of the inner palladium layer is not adversely affected by such irradiation. The uniformity of activity along the length of the radioactive sources irradiated with gamma radiation was confirmed by the optical density values of an exposed X-ray film and variations in optical density were found to be within \pm 6.2% (Fig. 3). These variations are very close to the deviations (i.e., \pm 5.8%) that are generally found in optical density values obtained with unirradiated ⁵I-sources.

Conclusion

¹²⁵I-brachytherapy sources could be fabricated by a previously optimized procedure by adsorption of ¹²⁵I on palladium coated silver rods. Individual sources containing up to \sim (74–111) MBq (2–3 mCi) of ¹²⁵I were uniformly coated with a thin coating of polystyrene. SEM micrographs of inactive samples of palladium coated silver rods irradiated with a gamma dose of \sim (17.85 × 10⁴) Gy (17.85 MRad), though revealed



Fig. 3. Optical density variations of gamma irradiated ¹²⁵I-source.

a mild agglomeration on the surface, the uniformity in the texture of palladium on the surface was maintained and subsequent adsorption of ¹²⁵I on irradiated samples was not adversely affected. Uniformity of activity on the irradiated radioactive sources was also found to be satisfactory. The leachability of the unirradiated as well as irradiated polystyrene coated radioactive sources was found to be well within permissible levels. The sources were found to be stable after absorption of radiation dose of ~(17.85 × 10⁴) Gy (17.85 MRad), and therefore can be repeatedly used for brachytherapy procedures over their practically useful life of about three weeks.

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