An approach to the preparation of iodine-I25 seed-type sources

Izabela Cieszykowska,
Andrzej Piasecki,
Mieczysław Mielcarski

Abstract The deposition of iodine-125 on silver bars to be sealed in titanium capsules as seed-type sources for brachytherapy was studied. For this purpose, a method of internal electrolysis was applied. The cell consisted of a silver anode contained in a Pt crucible serving as cathode. The plating solution contained sodium iodide-125 and sodium hydroxide. The yield of iodine-125 deposition for carrier-containing and carrier-free electrolyte solutions was determined. The deposition on uncutted Ag wires and on those cutted into 3 mm long bars was investigated. The influence of visible light on the coating process was examined. The leachability of iodine-125 from the silver wires as well as the activity distribution on the wires was measured. The obtained results indicate that the silver wire should be first coated with iodine-125 and then cutted into 3 mm long pieces. Based on this observation, a device for a batch-type procedure of manufacturing I-125 plated silver bars was designed and made. A design for simultaneous cutting and positioning of the activity-carrying bars into the titanium capsules is presented. This arrangement has been built and tested in trials. The tests confirmed that contamination of the source capsules can be minimized. Laser weldings of the titanium capsules were performed and the cross section of the welds examined by the use of an optical microscope.

Key words iodine-125 sealed sources • seed-type sources • brachytherapy sources

I. Cieszykowska, A. Piasecki, M. Mielcarski
Radioisotope Centre POLATOM,
05-400 Otwock/Świerk, Poland,
Tel.: +48 22/ 718 07 93, Fax: +48 22/ 718 03 50,
E-mail: m.mielcarski@polatom.pl

Introduction

Iodine-125 fixed on silver bars make a source core which, after being sealed in titanium capsules, are recognized as the so-called seed sources used in radiation therapy for treatment of intraocular and brain tumors, and as permanent implants for prostate cancer. The commercial manufacturers apply different methods for preparing source cores such as, for example, adsorption on organic materials, ion-exchange resins, ceramic beads, palladium bars, silver coated beads etc. Although some results of investigations concerning fixing of iodine-125 on silver are reported [4, 5], little technical information on the methods of source preparation is available. More details are available as concerns the dosimetric characterisation of commercially produced ^{125}I brachyseeds [1, 6, 9, 11–14].

The purpose of this work comprises an investigation of different parameters influencing the deposition of ^{125}I on a silver support in an electrochemical process occurring without the use of external electric power source, determination of leachability and uniformity of radioactivity distribution, and sealing thus obtained source cores in titanium capsules by laser welding.

The first step in developing a method for manufacturing iodine-125 seeds was the preparation of radioactive

source core. From the point of view of preparation of brachytherapy radiation sources, it seemed most practical if the deposition of iodine-125 occurred on a silver bar which acts as an X-ray marker. For this purpose, a convenient method of internal electrolysis was applied [7–10]. (This term indicates that the electrolysis of a solution occurs without the application of an external electric potential.) Such procedure is very simple, particularly as regards the experimental requirements which are of importance when radioactive electrolytes are used.

Experimental

The experimental techniques applied comprised making of radioactive source cores 3 mm long and 0.5 mm in diameter, positioning of the source cores inside the titanium capsules and welding the capsules.

Fixing of iodine-125 on silver wires

The deposition of iodine-125 on cutted and uncutted silver wires was studied. In the case of cutted silver bars, they must be removed from the plating solution after coating with ^{125}I and rinsed with water and/or aceton. Because of the small dimmensions of the cores such operation can cause problems as the cores require additional protection with a kind of lacquer. It may lead to sticking the pieces together after drying. The uncutted wire can be easily protected after coating with, e.g., polistyrene lacquer and the process of subsequent cutting and positioning of the cores in the titanium capsules can be done in a single operation.

In these experiments, 10 ml of electrolyte solution containing 0.01 M/l NaOH and either carrier-added or carrier-free iodine-125 was used. The activity of the plating solution was adjusted according to the estimated surface of the silver anode used. An assumption was made that the total activity fixed on a single silver bar, 3 mm long and 0.5 mm in diameter, should be close to 1.11 GBq. The coating solution was placed in a Pt crucible which served as cathode. The silver bars cutted into 3 mm long pieces were placed directly on the bottom of the crucible whereas the uncutted wires were immersed into the solution contacting the brim and the bottom of the crucible. Permanent stirring of the plating solution was not applied in spite of the duration of the deposition process. Sporadically, the cutted bars were stirred manually. The influence of light on the deposition of ^{125}I on cutted bars was investigated using UV-Mix Lamp 300 W (Śląska Fabryka Lamp Żarowych, Poland).

The deposition yield was determined by an indirect method. The residual non-adsorbed radioactivity in the plating solution and in the washings of the wires was measured using ionization chamber KG 4Π-50 with Keithley Electrometer type 485 (USA). For calibration of the system, standard solution supplied by the Holder of the National Standard of Radioactivity in Poland was used. The accuracy of the measurements was ± 5%.

Determination of the leachability of ^{125}I fixed on silver bars

Two pieces of the active wire, coated with a protective layer of polistyrene lacquer and one piece of unprotected silver wire, each 10 mm long, corresponding to 3 source cores, with ^{125}I activity in the range of ~1 mCi were placed in 5 ml of water at ambient temperature for 48 hours, according to PN-ISO 9978 [2]. The radioactivity of water was measured by the use of a scintillation system with a NaI(Tl) crystal and also with an X-gamma spectrometer with an HPGe detector (DSA-2000, GX-1820 manufactured by CANBERRA). The measurement systems were calibrated using a source prepared from the standard solution supplied by the Holder of the National Standard of Radioactivity in Poland.

Determination of uniformity of adsorbed ^{125}I on silver wires

The sources prepared by two different methods were checked for uniformity of the activity distribution. In the first method, similar to that described before, the silver wire was immersed slantwise in the plating solution contained in a Pt crucible, contacting the brim of the crucible above the solution level and also above its bottom. The obtained active Ag wires were protected with polistyrene lacquer. According to the second method, not applied in these investigations of fixing, the Ag bar was immersed in central position against the walls of Pt crucible without contacting its bottom. These active wires remain unprotected. In each method, the length of the active part was 10 mm.

Distribution of I-125 along the wire, 0.5 mm in diameter, was measured using a gamma counter equiped with a 1 mm collimator head. The energy window was set at 35.5 keV.

Encapsulation of activity-coated silver bars

This process include positioning of the activity-carrying silver bars inside the titanium capsules with one tip previously sealed. It should be performed in such a way as to avoid contamination of the capsules during loading. After checking the position of the active core, the second tip of the capsule is welded.

Laser welding of titanium capsules

For the source core encapsulation, a pulsed Nd:YAG laser welding system CTL-1508, produced by Laserinstruments Ltd. PL was applied. The main technical parameters of this welder are: wavelength 1064 nm; average power 50 W: pulse energy 50 J; pulse duration time 0.1–20 ms; pulse repetition frequency 1–10 Hz. Additional accessories comprised a CCD camera for observation of the welding process on a TV monitor and a mechanical system for precise positioning of the welded element against the laser beam.

Two different methods for laser welding of titanium capsules were examined. In both cases the laser beam was coaxial with the welded tube.

In the first method, the open tip of titanium tube contained a Ti plug. Rotation perpendicular to the long axis of the welded element was assured. The beam parameters were: power 4.5 W, pulse duration time 2 ms, pulse frequency 1 Hz. 38 beam pulses were applied per one turn of the welded capsule.

In the second method studied, the tip of the tube was squeezed and the welding was performed by applying a single 5 ms pulse of 6 W power.

The cross section of the capsules were visualised by the use of an optical microscope (enlargement × 200) after grinding and etching.

Results and discussion

Fixing of iodine-125 on silver

The current-voltage characteristics of the cell used in the internal electrolysis process for fixing iodine on silver were described earlier [7, 10].

The effect of carrier iodide concentration and time on the yield of the adsorption of I-125 on uncutted silver wires is shown in Fig. 1. It can be seen that the decrease in activity of the plating solution slightly depends on the carrier concentration. The process runs little faster when the carrier concentration is higher, although the difference can be considered as negligible. The deposition yield of fixing iodine-125 on uncutted silver wires from both carrier-added and carrier-free solution is shown in Fig. 2. In the case of carrier-free solution, the adsorption proceeds more slowly, but the total yield remains the same and amounts to 97%.

The results obtained for cutted 3 mm long silver bars, immersed directly at the bottom of a Pt crucible containing carrier-free ^{125}I solution are shown in Fig. 3. The deposition yield on cutted bars was considerably lower than that on the uncutted ones. This can be caused by an increase of electric resistance between the Pt crucible and Ag bars coated with AgI layer whose conductivity is lower than that of silver. One can expect that the thicker silver iodide layers obtained from the carrier containing solution could cause even higher
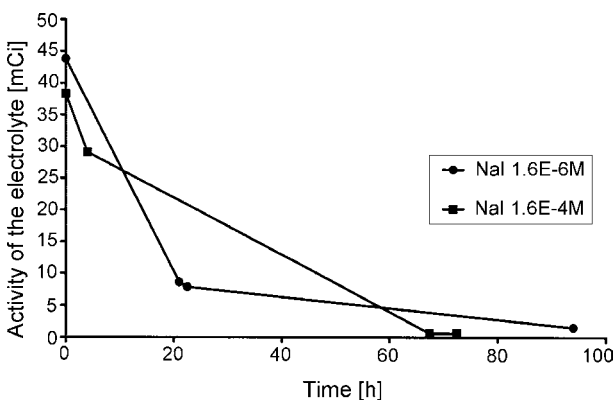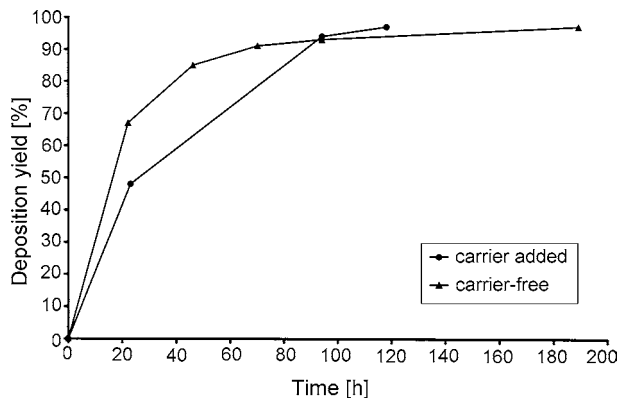

Fig. 2. Deposition yield *vs.* time for carrier-added and carrier-free solution.
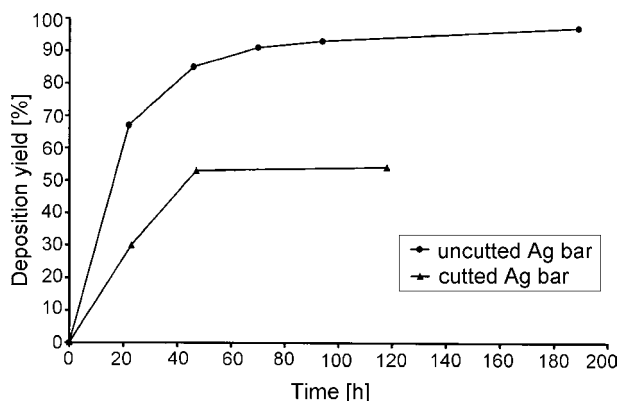

Fig. 3. Deposition yield on cutted and uncutted silver bars.

resistance and further decrease of the deposition yield. This is confirmed by the results plotted in Fig. 4. The presence of the carrier reduces the deposition yield from 54% to 29%.

As it follows from the literature data [15], the electric conductivity of AgI increases under the influence of light. Therefore, it could be expected that the deposition yield should also increase when the solution is illuminated. This is confirmed by the results shown in Fig. 5. Similar effect was not observed in the case of deposition on uncutted bars where the electric contact between Pt crucible and bare silver wire is assured outside the plating solution (Fig. 6).
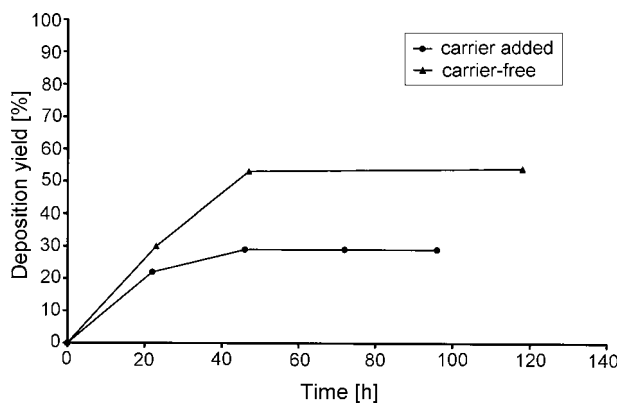

Fig. 1. Effect of carrier iodide concentration and time on the adsorption of ^{125}I on silver.

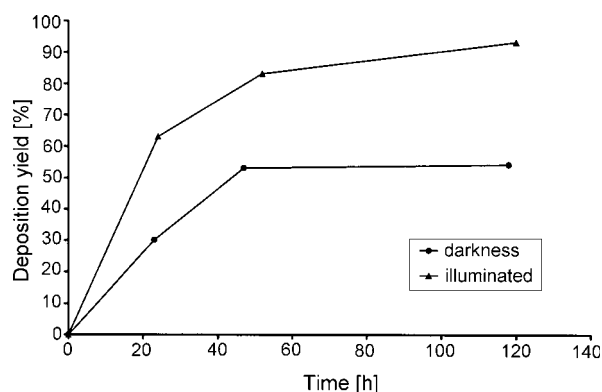**Fig. 4.** Deposition yield on cutted Ag bars from carrier added and carrier free ^{125}I solutions.

Fig. 5. Effect of illumination on deposition yield of ^{125}I on cutted silver bars.
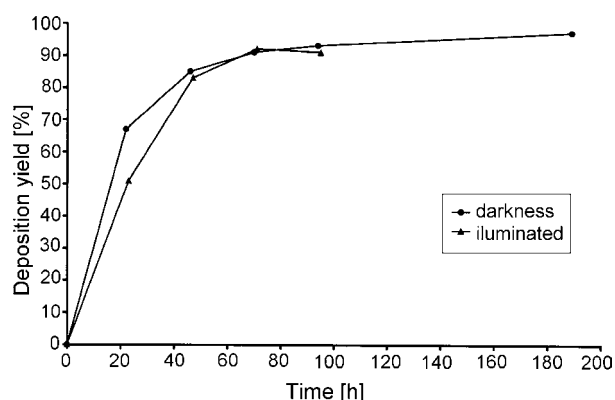

Fig. 6. Effect of illumination on deposition yield of ^{125}I on uncutted silver bars.

The photoelectric effect on the conductivity of silver chloride, bromide and iodide was studied earlier (by W. Wilson) [15]. His measurements of the conductivities of binary mixtures of silver chloride, bromide and iodide showed that the conductivity is raised if the salt is exposed to iodine vapour. Besides, it can be expected that illumination causes reduction of AgI to metallic silver causing an increase of the conductivity enhancing the plating process. The released ^{125}I migrates back to the alkaline solution, whereas the reduced Ag forms a porous layer on the silver anode. On the expanded anode surface additional adsorption of radioiodine can occur. Therefore, electrodeposition and adsorption can be considered as two parallel processes.

Determination of the leachability of ^{125}I fixed on silver bars

The results of the leachability are presented in Table 1.

Table 1. Leachability of ^{125}I wire sources.

Description	Activity [MBq]	Activity leached out [kBq]/[%]
Protected Ag wire	35.43*	15.3*/0.04
Protected Ag wire	40.00*	43.1*/0.1
Bare Ag wire	32.28*	608.2*/1.9

   * Uncertainty of activity determinations ±5%.

Single samples of each kind were examined. The leachability of ^{125}I from the protected silver bars was very low. In case of the unprotected source it was higher although still lower than other results reported [5].

Determination of activity distribution on silver wires

The activity distributions along the protected and unprotected silver wires are shown in Figs. 7 and 8, respectively. The observed deviation from uniformity results probably from the technique of coating. In the simplified method related to the protected bar, the silver wire was immersed aslant in the plating solution contacting both the brim and the bottom of the Pt crucible. These two unequivalent contact points between the silver anode and Pt cathode could cause disturbances in the current flow during the plating process. Better uniformity was obtained for the unprotected silver wire which was immersed in the plating solution vertically with one contact point with the Pt cathode. However, also in this case better uniformity is observed for the central parts of the wire than for its ends. Similar effect was observed by Kuznetsov *et al.* [3] for palladium coated nickel wires.

Batch method for manufacturing source cores

Based on the obtained results, it has been decided that the technique applied for a batch type manufacturing should comprise fixing first and afterwards cutting the
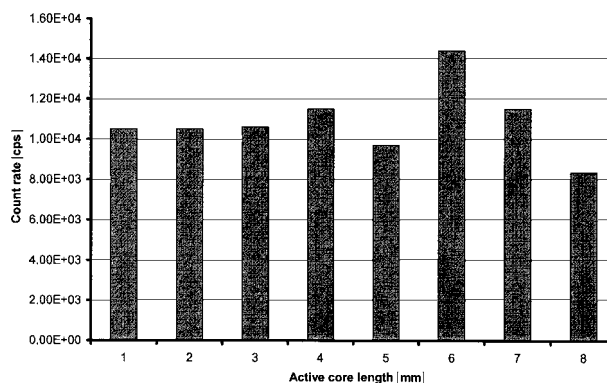

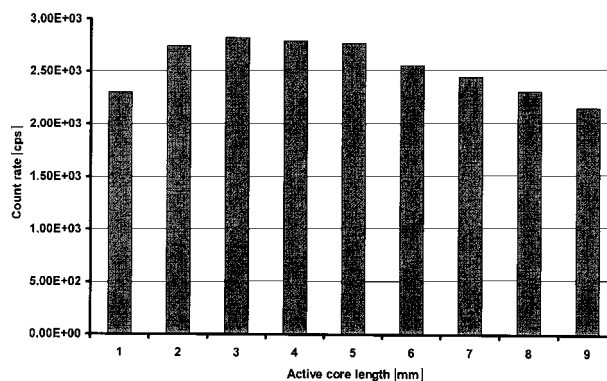**Fig. 7.** Activity distribution on protected silver wire.

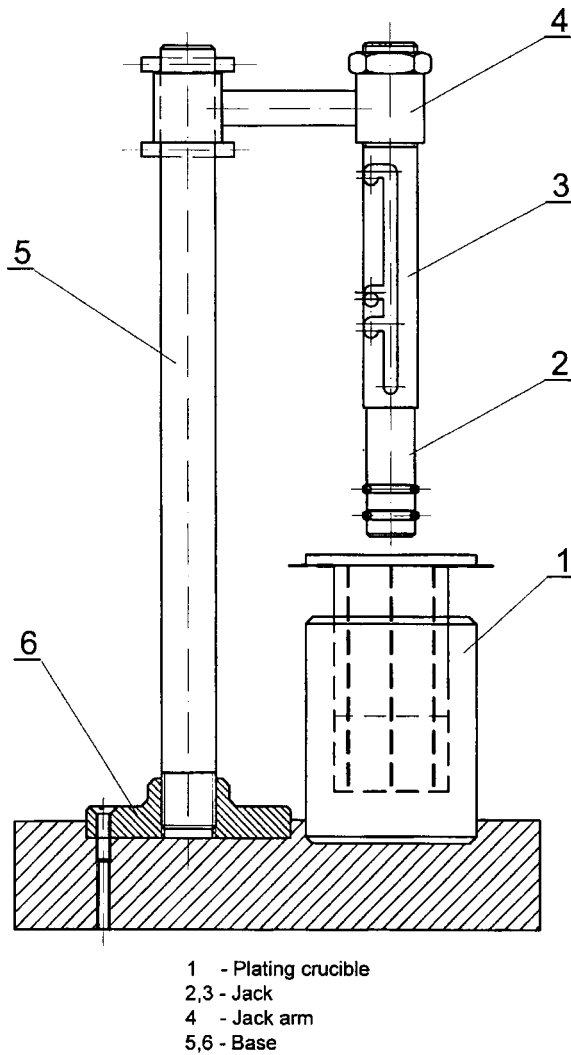**Fig. 8.** Activity distribution on unprotected silver wire.

1   - Plating crucible
2,3 - Jack
4   - Jack arm
5,6 - Base

Fig. 9. Plating assembly.

1   - Rotary disk
2,3 - Fixed disks
4   - Distance sleevs
5   - Axis of rotation
6   - Base
7,8 - Cutting elements
9   - Ag wire
10 - Positioning device

Fig. 10. Cutting and positioning device.

Laser welding of titanium capsules

The cross sectional view of the weld obtained according to the first method described in the experimental part is shown in Fig. 11a and must be considered as unsatisfactory.

The result obtained by the second method is picturized in Fig. 11b. The shape of the obtained weld is of good quality although the cross sectional view shows some unevenness of the melted metal.

a   b

Fig. 11. Laser welds of titanium tubes (a) with plugged tip; (b) with squeezed tip.

active silver wire into 3 mm long pieces. Following this conception, a device for a batch-type preparation of silver bars coated with I-125 was designed and made. This arrangement, shown in Fig. 9, enables preparation of 4 active bars, each 10 mm long, in a single procedure although the number can be increased according to demands. The central hole in the blank holder and wire holder enables one to collect samples of the plating solution for activity measurements during the coating process.

After completion of the plating process, the silver wires are removed from the plating solution and allowed to dry. For additional protection they can be immersed in a container with 3% solution of polystyrene in chloroethylene.

Cutting and positioning of active silver bars inside the titanium capsules

The design for simultaneous cutting and positioning of the activity-coated silver bars inside the titanium capsules is shown in Fig. 10. This arrangement allows one to avoid contamination of the capsules during loading which has been confirmed in several trials.
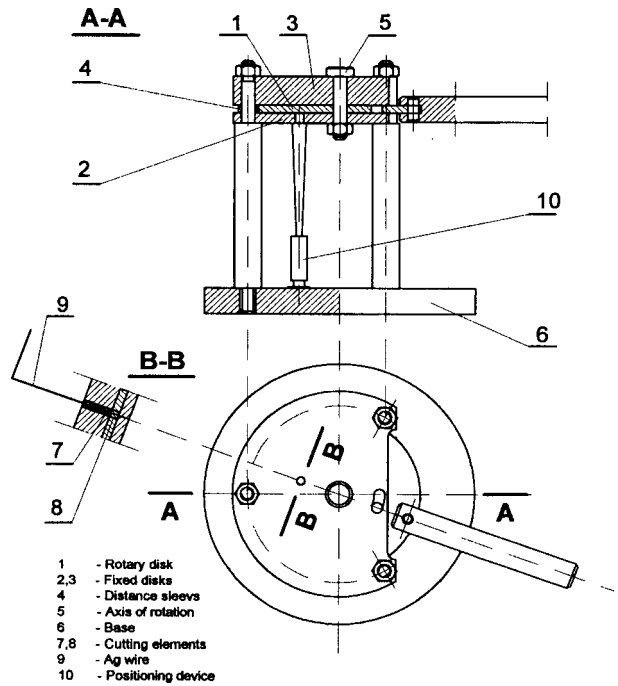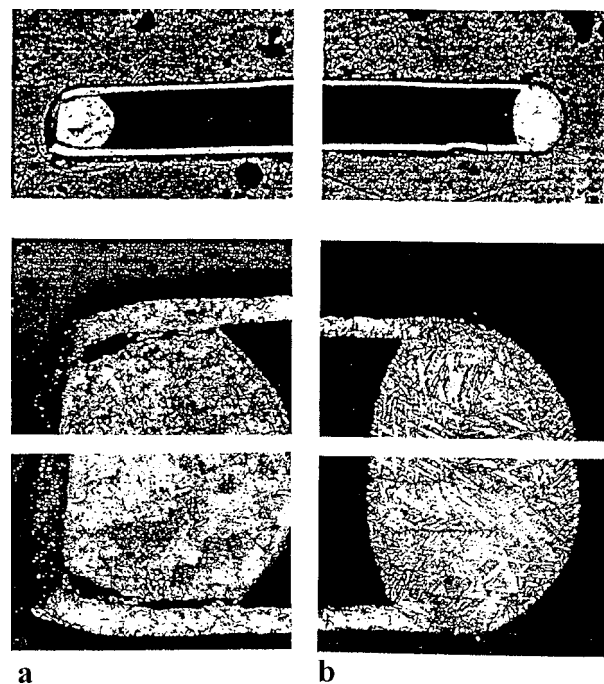
Our recent experiments, in which different welding parameters were further adjusted, showed that the quality of the welds can be improved.

Conclusion

Among the various methods of fixing of iodine-125 on silver wire, internal electrolysis is simple and effective. The experimental conditions ensure safe handling of high amounts of radioactivity without the release of iodine-125 to the air. The coated Ag bars require careful handling for encapsulation. This is assured by the designed equipment. The latest experiments on laser welding showed that the quality of welds can be improved. For medical applications, the encapsulated sources will have to be assessed by stringent quality control tests such as surface contamination, leak tests and activity measurements.

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