IAEA-TECDOC-1512

Production techniques and quality control of sealed radioactive sources of palladium-103, iodine-125, iridium-192 and ytterbium-169

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PRODUCTION TECHNIQUES AND QUALITY CONTROL OF SEALED RADIOACTIVE SOURCES OF PALLADIUM-103, IODINE-125, IRIDIUM-192 AND YTTERBIUM-169

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FOREWORD

Radioisotopes have been used extensively for many years for several medical and industrial applications either in the form of an open source or encapsulated in an appropriate metallic container (sealed source). The design and technology for the preparation of radioactive sealed sources is an area of continuous development to satisfy an ever increasing demand for a larger variety of shapes, sizes, type of radioisotope and levels of radioactivity required for newer and specialized applications. In medicine, sealed sources using the radioisotopes of ^{125}I, ^{192}Ir and ^{103}Pd are commonly used for brachytherapy for the treatment of malignant diseases, and for bone density measurements. In industry, they are widely used for non-destructive testing (NDT), radiation processing, "on-line" process control systems and on-line elemental analysis of mineral resources. Some well-known examples of such sources are ^{60}Co for industrial nucleonic gauges, ^{192}Ir sources for industrial radiography, ^{241}Am sources for smoke detectors and chemical analysers and, more recently, ^{169}Yb for NDT measurements of thin metallic tubes and plates. The current challenges in development include the production of miniature size sources with a high level of activity, a high degree of uniformity in the distribution of the radioactivity and the highest degree of safety, requiring stringent quality control methods.

The IAEA has been promoting and supporting activities designed to increase the utilization of radiation and radioisotopes in several areas. In particular, in view of the proven benefits of, and an increasing demand for radioactive sealed sources for medical and industrial applications, upon the recommendation of several experts, a Coordinated Research Project (CRP) on Development of Radioactive Sources for Emerging Therapeutic and Industrial Applications was begun in 2002. The aim of the CRP was the optimization and testing of procedures and methods for the fabrication and quality control of sealed sources based on Iodine-125, Palladium-103, Iridium-192 and Ytterbium-169. Experienced scientist groups from Belarus, China, Hungary, India, the Islamic Republic of Iran, Kazakhstan, the Republic of Korea, Peru, Poland and the Russian Federation participated in the CRP under research contracts and agreements.

The technology and experimental procedures described in this report are the result of the common collaborative research of all the participants in the CRP. Many of these procedures are innovative and yet simple to follow by anyone wishing to prepare radioactive sealed sources based on ^{125}I, ^{103}Pd, ^{169}Yb and ^{192}Ir.

The IAEA wishes to thank all the participants in the CRP for their valuable contributions. The IAEA officer responsible for the CRP and this report was M. Haji-Saeid of the Division of Physical and Chemical Sciences.

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SUMMARY

1. INTRODUCTION

Radioactive sealed sources have been used since several years for a wide range of applications in a variety of shapes, sizes and radioactivity levels. In industry, they are widely used for non-destructive testing, radiation processing, “on-line” process control systems, on-line elemental analysis for raw materials, semi-formed and final products, mineral resource evaluation, food irradiation and smoke detection. In medicine, they are commonly used in teletherapy and brachytherapy for the treatment of malignant diseases, and for bone density measurements. In research, a variety of sources for different applications are used, most commonly for elemental analysis (i.e. X ray and neutron activation analysis) and material structure studies. Some well-known examples of such sources are ^{60}Co sources for teletherapy, brachytherapy, food irradiation and sterilization of single use medical supplies, as well as industrial nucleonic gauges, ^{192}Ir sources for industrial radiography and brachytherapy, ^{241}Am sources for smoke detectors and ^{137}Cs sources for brachytherapy.

The activity intensities of these sources range from a few μCi (1 Ci = 3.7x10^{10} Bq), used for calibration purposes, to several thousand curies in industrial irradiators and radiation therapy. The facilities for the fabrication of these sources vary in complexity, depending on the type of radiation, for instance, X rays, gamma rays or neutrons, and the levels of radioactivity handled. The radioactive isotopes are used in a variety of chemical and physical forms, metallic or oxide forms, impregnated into ceramics, electroplated on other support metals as thin films or deposits. They are encapsulated into inert metallic capsules in many cases as sealed radioactive sources.

Radionuclides commonly used for industrial sealed sources are ^{60}Co, ^{63}Ni, ^{90}Sr, ^{137}Cs, ^{147}Pm, ^{169}Yb, ^{170}Tm, ^{192}Ir, ^{241}Am and others. The demand and scale of industrial applications of sealed sources for some of these sources like ^{137}Cs has decreased dramatically in the past few years due to safety concerns. The demand for ^{60}Co seems to be nearly stable, whereas the demand for ^{192}Ir, on the other hand, is expected to increase in the next few years. One of the recent developments in this area is the use of ^{75}Se-based sources for non-destructive testing, for which the demand is also expected to increase in the next few years, due to the fact that it provides images of higher quality in many cases.

During the last decade the most important developments in sealed sources have been related to medical applications. In contrast to the earlier situation of using a limited number of radionuclides, mainly ^{60}Co and ^{137}Cs, the present scenario offers a wider range of radionuclides including ^{125}I, ^{103}Pd, ^{90}Y, ^{90}Sr, ^{144}Ce and ^{106}Ru, among others. The most prominent recent developments in sealed sources include ^{192}Ir sources for high-dose rate brachytherapy, ^{125}I and ^{103}Pd seeds for the treatment of prostate and brain cancers, catheter mounted sources, stents and balloons incorporating ^{90}Sr, ^{90}Y, ^{144}Ce, ^{166}Ho, ^{32}P and ^{133}Xe for vascular therapy, ^{125}I and ^{106}Ru sources for ophthalmic applicators. ^{60}Co sources are used in “gamma-knives” for the treatment of brain tumours. Among these, the radioactive seeds for implantation into human tissues, sources for vascular therapy are of particular interest, in view of their potential new applications.

The miniature size of these sources and the high degree of uniformity in activity distribution needed present technical challenges in production and effective quality control. Many of these sources, such as ^{125}I, ^{103}Pd, ^{144}Ce and ^{169}Yb sources, are based on the active core production method from the primary radioactive preparations. Various techniques can potentially be applied to radionuclide deposition/fixation in the active core, such as electrochemical deposition, adsorption, precipitation, pressing, etc. These techniques can include special procedures for improving active core/source properties, such as decreasing radionuclide leaching in the case of unforeseen source damage. These are examples of potential areas for R&D efforts in source production.

The necessity of specially designed and remotely operated precise positioning systems for source assembling and sealing (welding) in a hot cell, as well as the automation of processes, offers another area for developmental efforts. Development of quality control methodologies is also an area of active investigation.

The International Atomic Energy Agency (IAEA) organized a Consultants Meeting in April 2001 in Vienna to review the current status, recent developments and future trends in radioisotope sealed sources. The consultants felt that, in view of the fact that sealed sources play a very significant role in radioisotope applications in several fields and, considering the promising new developments in this area, R&D activities in the preparation and quality control (QC) of sources deserve support. They recommended that development of production and QC methodologies of such miniature sources, with applications in therapy and selected applications in industry, still have unresolved technical problems that need further investigation, and can be an appropriate subject for a coordinated research project (CRP) of the IAEA.

Based on the consultants' recommendations, the IAEA has approved the implementation of a three-year long Coordinated Research Project entitled Development of Radioactive Sources for Emerging Therapeutic and Industrial Applications beginning in 2002. The CRP focused on the optimisation and testing of procedures and methods for the fabrication of sealed sources based on Iodine-125, Palladium-103, Iridium-192 and Ytterbium-169. Although the production of the primary radionuclide was not strictly considered a part of the CRP goals, for the particular case of ^{103}Pd, the investigation of its production in nuclear reactors was considered to be an important part of the CRP.

The main technical goals of the CRP as defined at the outset of the project are described as follows:

- investigation of methodologies for miniature source core production and development of technology for batch-type production of such sources (for example ^{125}I or ^{103}Pd seeds and high dose rate ^{192}Ir sources for brachytherapy),
- development of techniques/devices for assembling and sealing such sources, with emphasis on precise positioning, source core encapsulation and welding techniques and
- development of methods for quality control of such sources, including activity calibration and activity distribution measurement.

Experienced scientists from Belarus, China, Hungary, India, the Islamic Republic of Iran, Kazakhstan, the Republic of Korea, Peru, Poland and the Russian Federation participated in the CRP under research contracts and agreements. The 1st Research Coordination meeting (RCM) was held on the premises of the Radioisotope Centre POLATOM, in Otwock-Swierk, near Warsaw, Poland, 24–27 June 2002. The meeting discussed the importance of continuing R&D related to the production of miniature sealed sources for brachytherapy and industrial uses as well. The report resulting from this meeting outlined the specific goals of the CRP as reflected above. In particular, it was the common consensus of the participants that the problems to be investigated under this CRP would include: reduction of the production cost of the radionuclide of interest whenever possible, active source core preparation, positioning of the active core in a miniature source capsule, investigations of capsule welding techniques, reduction of contamination of the equipment and the environment in the manufacturing process, activity calibration and quality control.

The 2nd RCM that took place at the IAEA in Vienna, 20–24 October 2003 highlighted the progress achieved during the first period of the CRP. Improvements in source core preparation following a variety of methods were reported, giving fixation yields over 80-90% for ^{125}I and ^{103}Pd, depending on the method followed. In terms of source core quality, low leachability was demonstrated, paving the way for further steps like source encapsulation. In this regard, several welding techniques such as laser and plasma were investigated.

Moreover, areas for further research within the remaining period of the CRP were identified. In particular, it was decided to give emphasis to: a) investigation on radioisotope (^{125}I) fixation on different kinds of source core support (ceramic, ion exchange and metallic beads), b) optimisation of encapsulation and welding techniques (laser, micro-plasma and TIG welding), and c) development or improvement of existing QC procedures (welding quality, dosimetry and activity measurement).

At the third and final RCM, again held at the IAEA in Vienna, 13–17 June 2005, the final reports of the research agreements and research contract holders were presented, and their results were discussed and evaluated. The scientific results, as discussed below, indicate that the original goals of the CRP were essentially met. Details of the results and achievements of the project are given in the next section.

2. SUMMARY OF THE ACHIEVEMENTS OF THE CRP

In general, it was found that all participants involved in the development of the production technologies of miniature sealed sources have reached considerable progress, to the point that several key technological steps have been mastered and put into practice for the manufacturing of such radioactive sources for medical and industrial applications, as well. It is concluded also that communication and collaborative research among the participants was strengthened, and it is expected that these fruitful contacts will continue in the future. The examples of such collaboration include data exchange and detailed technical discussions among the participants during the research coordination meetings, distribution of the published materials, distribution of available technical information on experimental procedures such as for welding techniques, distribution of sample dummy capsules produced by the participants, as well as advice and recommendations of particular laboratory techniques proven to give better results.

The major technical and scientific developments achieved by the participants during the period covered by the CRP are summarized in the following sections.

2.1. Sources for medical applications

2.1.1. Iodine-125, Iridium-192 and Palladium-103

1. Various methods for fixing radioisotopes like ^{125}I and ^{103}Pd were developed for the preparation of radioactive source cores based on different matrices, such as silver and nickel wires and ceramic rods. The methods used for fixation of these isotopes were based on different techniques, such as electro-deposition, internal electrolysis, physical adsorption and chemical / physical adsorption. The fixation of the radioisotopes varied from 80-90%, depending on the method used. Thus, chemical-physical adsorption of efficiencies of 98% for ^{125}I in Peru, 83% in Iran and 94% in India were reported. By the electrodeposition technique, the participant from India obtained 84% efficiency. The participant from India reported fixation efficiencies for ^{125}I on the silver-containing alumina rods of 95-98% and 87% on metallic silver rods. The method of adsorption of iodine-125 on chlorinated silver bars developed in China gave an efficiency of about 90%. Following the internal electrolytic method the participant from Poland reported fixation efficiencies of 98%. The novel method of sublimation developed by the participant from Kazakhstan using ^{131}I as a tracer produced fixation yields of 95%.

2. Two relatively new approaches of iodine fixation on a solid matrix were investigated during the CRP implementation. They are dry distillation of iodine in vacuum, followed by its adsorption on a silver surface from gaseous phase, and chemo-sorption on a ceramic material modified by metal silver particles. These approaches provide higher matrix activity that can be used for large scale (batch type) production, including sources of irregular shapes.

3. The leachability of the source cores was examined by standard national protocols before encapsulation. The source cores prepared by various methods had low leachability and could be used for encapsulation: 0.01% for the sources prepared in Kazakhstan and India, 0.04 % in Poland, 0.06 in Iran and 0.2 % in Peru.

4. The source cores prepared were encapsulated in titanium or stainless steel capsules and welded. The welding was mostly carried out by laser for titanium capsules and by micro plasma and TIG welding for stainless steel ones. Electron beam welding of the sources was also carried out by one of the participating countries.

5. Laser welding parameters such as beam diameter, output power, optical system, etc., were optimized to obtain good quality welds which were confirmed by metallographic studies.

6. Quality control tests including examining various parts of the capsule before welding, leak tests to check welding, wipe tests for radioactive contamination, accurate activity measurements and source uniformity of the welded capsule were studied.

7. Clinical trials of the ^{125}I-sources were carried out in India in which five patients were treated for malignant melanoma and retinoblastoma. It is noteworthy that one patient who was treated for retinoblastoma after unsuccessful External Beam Radiation Therapy showed remarkable improvement, which is highly gratifying. It can be concluded that external beam radiotherapy often fails in the treatment of ocular tumours, which respond well to the brachytherapy treatment.

8. It was demonstrated that ^{103}Pd could be effectively produced by reactor (moderate and high flux) irradiation of enriched target of ^{102}Pd (>50%) providing ^{103}Pd with specific activities, sufficient for miniature source production, up to 500 Ci/g in a high-flux reactor (1 Ci = 3,7x10^{10} Bq).

9. ^{192}Ir high dose rate (HDR) sources (up to 12 Ci/source of 1.1 mm in diameter and 4.5-5.3 mm in length) are produced by sealing reactor irradiated pellets (0.6 mm dia x 3.5 mm length) or disks (0.5 mm Ø x 0.5 mm height) in stainless steel capsules pre-joined with the guiding rope. Laser welding is used for both source capsule sealing, as well as capsule-rope joint.

10. Welding conditions were optimized to ensure ^{192}Ir sources classification C53211 according to ISO 2919:1999 standard.

11. For intracavitary brachytherapy large-size sources of ^{192}Ir (4.6 mm outer diameter and 6.5 mm length) connected to a flexible cable can be used. In this case tungsten inert gas (TIG) welding can be applied for source fabrication.

12. Clinical trials with ^{125}I seeds and ^{192}Ir sources were reported by several participants. The initial results of these trials show encouraging prospects for use in the treatment of some forms of cancer.

2.2. Sources for industrial applications

2.2.1. Ytterbium-169

1. A point source based on ^{169}Yb was produced by a high flux reactor using the ^{168}Yb(n,γ) nuclear reaction. Since natural ^{168}Yb has an abundance of 0.13%, enriched targets (>20% enrichment) were used to attain proper specific activity.

2. Optimal compacting and sintering conditions of Yb_2O_3 powder were determined experimentally to fabricate miniature pellets of 1x1 mm.

3. To ensure "clean" conditions for final source assembly after irradiation, pellets were encapsulated in aluminium capsules and then irradiated in a reactor to produce activities of 5-10 Ci/pellet.

4. The irradiated aluminium capsules with ytterbium pellets are sealed in outer titanium capsules to ensure a higher degree of safety. Besides, the outer capsule provides the means to connect the source to a rope-type holder.

5. A source holder for ^{169}Yb sources, which can fit in conventional NDT projectors, was designed.

2.2.2. Iridium-192

1. The assembling and welding techniques of ^{192}Ir sealed sources intended for industrial application were optimized. An active core of this kind of source is a set of irradiated iridium disks with a diameter of 0.5 to 3.5 mm and a thickness of 0.2 to 0.5 mm (depending on required source activity and dimensions).

2. Specific activities from 250 to 400 Ci/g ($9.25\ 10^3 – 1.48\ 10^4$ GBq/g) of ^{192}Ir were achieved by irradiation in moderate to high flux neutron research reactors of metal Iridium with natural isotopic composition and were found to be satisfactory for the fabrication of miniature ^{192}Ir industrial sources.

3. Capsules made of stainless steel and sealed by laser or TIG welding were successfully double encapsulated for safety reasons. This design provides a source classification C43515 according to ISO 2919:1999 standard.

3. OUTPUTS AND OUTCOMES OF THE CRP

Updated information and know-how for the production and quality control of sealed radiation sources were discussed in the initial phases of the CRP, including issues related to unresolved technical problems of the involved technology. The results of the investigations of these problems through the CRP have generated very useful data of interest to laboratories in several Member States. In addition, the participants have improved or developed new methods of production, testing, QC methodologies and encapsulation technology of sealed sources for applications in medicine and industry. Several results of the CRP are worth mentioning. All participants not only adopted the existing technology but also developed new technology to satisfy the application envisaged. The radioisotopes used for the development of sealed sources were ^{125}I, ^{192}Ir, ^{103}Pd, and ^{169}Yb.

^{125}I has been used for the preparation of miniature brachytherapy sources by all the participants except the Russian Federation who have used ^{103}Pd as source core material. The primary radioisotope was purchased by all the countries except India and Korea who have their own indigenous supply. The radioisotope of ^{103}Pd was produced in Russia by neutron irradiation of enriched ^{102}Pd in a high flux nuclear reactor.

3.1. Sources for medical applications

3.1.1. ^{125}I sources

A novel approach for the adsorption of ^{125}I on silver wire as a matrix for the source core has been developed by India and adopted by Iran and Peru. Physical adsorption of ^{125}I activity on the $PdCl_2$ treated silver wire was suitable for adsorption. It offers the following advantages:

- easy operation/process,
- quantitative ^{125}I adsorption,
- availability of silver beads/wires with high purity,
- non-leachability of ^{125}I activity from the source core,
- mild experimental conditions of adsorption at neutral to alkaline pH,
- safe handling of high amounts of radioactivity and
- flexibility in bath size.

As a result of their participation in the CRP, three countries managed to produce ^{125}I source core for therapeutic applications. The CRP thus demonstrated that data exchange and detailed technical exchange among the participants help others to build their capability on a small but relevant scale.

A new technique for the adsorption of ^{125}I on chlorinated silver bars was developed in China and adopted for making source cores. Source core activities up to 1 mCi (37 MBq) have been prepared with low leachability. Further work is in progress.

A new and innovative sublimation approach has been devised and tested for the preparation of source core (Kazakhstan) based on the principle of thermal sublimation of free iodine in vacuum and subsequent chemosorption on substrates. The unique advantage of this method is that sources of various shapes and sizes can be prepared and activities up to 100 mCi (3.7 GBq) could be deposited on the core of standard brachytherapy seeds with very high uniformity (±2% deviation). This approach can be extrapolated for the preparation of sources of higher strength for their potential application in lixiscopy and bone densitometry. Another efficient but simple method based on the absorption of ^{125}I in a ceramic matrix treated with silver nanoparticles was investigated (Korea).

A very efficient but simple internal electrolysis technique has been developed in Poland, automated and implemented under this CRP. The advantages of this method are:

- it is a straightforward procedure,
- the degree of deposition can be adjusted by controlling the current,
- the non-leachability of ^{125}I activity,
- the automatic closed system allows safe handling of large amounts of radioactivity and
- there are no / less maintenance problems.

An indigenous technology for the regular production of ^{125}I has been tested and developed by the Indian participant to meet the in-house requirement and possible export. The technology refers to the production of spherical ^{125}I-beads on account that relatively higher dose rate delivery may reduce the total treatment time and have the potential to replace the presently used rod type ^{125}I-sources in eye and prostate brachytherapy. These sources have undergone clinical trials in India in which 5 patients were treated. Out of these five cases, two cases were of retinoblastoma and three cases of malignant melanoma.

3.1.2. ^{103}Pd sources

Under the CRP, methods for the production of ^{103}Pd and seed cores of palladium have been developed by the Russian Federation participant.

An approach based on nuclear reactor irradiation of isotopically enriched ^{102}Pd (up to 80%) has been tested for ^{103}Pd production. Irradiation in the high-flux reactor (neutron flux density higher than $1x10^{15}$ $cm^{-2}s^{-1}$) provides palladium-103 with specific activity up to approx. 500 Ci/g ($1.85\ 10^{4}$ GBq/g). Based on this result, it can be envisaged that moderate flux reactors with neutron flux density of 10^{14} $cm^{-2}s^{-1}$ can be also used for ^{103}Pd production with specific activities much higher than the required minimum of 5 Ci/g ($1.85x10^{2}$ GBq/g) for brachytherapy seed production.

Advantages of reactor production are the possibility for large-scale production of ^{103}Pd, simplicity of target preparation and post-irradiation procedures. The experience gained by the Russian participant under the CRP can be easily transferred to any group interested in the production of ^{103}Pd sources.

An electrochemical procedure has been developed for nickel wire plating with ^{103}Pd from phosphate type electrolyte consisting of palladium chloride, ammonium and sodium phosphate and ammonium chloride. Advantages of this electrolyte are as follows:

- production of uniform palladium layers with good adhesion,
- simple preparation,
- stability of the composition during electrolysis and the possibility to correct composition if required, and
- minimal losses of palladium-103.

A procedure for the preparation of nickel wire support before palladium plating has been developed to ensure homogeneous distribution of palladium along the wire. It is based on sintering in vacuum and washing in a mixture of hydrochloric and nitric acids (1-2 min at ambient temperature or 30 sec at 50^{o}C) followed by washing and rinsing with water and ethanol.

It also has been demonstrated that an electrochemical cell with flat anode and multiple cathodes can be used for batch type production of seed cores. Uniformity of palladium distribution better than 95% was demonstrated by ^{103}Pd activity measurements in axial and radial directions using a collimator technique.

The procedure of ^{103}Pd seed core production developed under the CRP can be applied to both reactor and cyclotron produced ^{103}Pd, so any person with access to radioactive ^{103}Pd can apply it for seed core production.

Calibration factors were also determined, allowing the evaluation of palladium radiation absorption by a titanium capsule, the palladium layer and nickel rod. This data is relevant not only for the development of ^{103}Pd seed production technology but also for ^{125}I seeds.

3.1.3. ^{192}Ir Brachytherapy sources

The participants found that laser-welding methods are in general preferable for miniature source encapsulation. QC methods were developed according to the corresponding ISO standards.

The participants from Belarus and Korea have already started the batch manufacture of brachytherapy sources based on the technology described in this report. They have already received the Special Form Radioactive Material Approval Certificate from the relevant national authorities. A few brachytherapy sources were already installed and used in after loading machines.

3.2. Sources for industrial applications

3.2.1. Iridium-192

The participant from Belarus, who has already received the Special Form Radioactive Material Approval Certificate, has already started the batch manufacture of industrial ^{192}Ir sources based on the technology described in this report. A few dozen sources have already been installed and used in the NDT projectors.

3.2.2. Ytterbium-169

The participant from Korea has successfully developed the technology for the preparation of miniature ^{169}Yb sources for industrial applications (NDT testing). The salient points of this development are as follows:

- a technology for large scale production of Yb_2O_3 pellet production (of 1 mm in diameter and 1 mm in height) was developed based on pressing at 90-100 kg, followed by sintering at 1050^{o}C,
- techniques for pellet encapsulation in aluminium capsule before irradiation and in titanium capsule after irradiation (by laser welding) have been developed and reported, thus making them available to others interested in the production of these kinds of sources and
- sources of the suggested design can be used for substitution of ^{192}Ir sources applied in conventional NDT projectors, providing better quality of the radiographic pattern.

The Korean participant is ready to produce ^{169}Yb sources and to supply them to customers in industry.

4. PROCEDURES AND METHODS

4.1. Iodine-125

4.1.1. Primary radioisotope production

^{125}I is routinely produced by neutron irradiation of ^{124}Xe gas. This isotope is supplied by many enriched isotope-producing companies on a regular commercial basis. Most of the CRP participants purchased ^{125}I from commercial producers for developing ^{125}I sources production technology. However, India and Korea have their own supply of these isotopes. In India, ^{125}I is produced from the neutron irradiation of natural xenon gas target using the wet chemical process, whereas Korea produced ^{125}I from the irradiation of enriched Xe target, employing the dry distillation method.

4.1.2. *Technical requirement of the source*

The specifications for sealed sources for brachytherapy are:

- high radionuclide purity of the isotope i.e.^{125}I,
- activity of source should be ± 10%,
- repeatability of dimension of seed-type sources after encapsulation and
- leak tightness of sources.

The corresponding major requirements for the source core are:

- good uptake of ^{125}I into a solid matrix,
- uniform distribution of ^{125}I activity in the support substrate,
- low leachability,
- product reproducibility within acceptable tolerances and
- processes for source preparation in large batch sizes should be feasible in a minimum amount of time.

4.1.3. Source core preparation

The ^{125}I source core usually consists of silver backing, which acts as an X ray marker, with the radionuclide activity fixed on its surface. In some cases a ceramic support is used. The main challenge, therefore, was to develop a non-leachable source core containing ^{125}I. Production of the source core in a highly reproducible manner within acceptable dimensional tolerances was yet another challenge. Some innovative strategies have to be adopted to develop the necessary methodology for fabrication of ^{125}I source cores, capsules and a reliable and reproducible encapsulation technique. Some of the techniques and matrices used for making ^{125}I source core are electrochemical deposition, internal electrolysis, adsorption, sublimation, absorption on porous glass or ceramics etc.

4.1.4. Physico-chemical adsorption

Pure silver wires (0.5 mm diameter) cut into pieces of 3 mm length were thoroughly washed with acetone, followed by hot and cold water rinsing. They were further treated with 3 M HCl and washed free of acid with double distilled water, were completely dried under an IR lamp to a constant weight and were used for adsorption experiments. The pre washed silver wires were immersed in $PdCl_2$ solution at acidic pH. The solution was boiled for 5 min and cooled. The wires were thoroughly washed in double distilled water and dried to a constant weight. The cleaned wires were individually immersed in small conical shaped test tubes containing a mixture of ~148 MBq (~ 4 mCi) of ^{125}I and 5 µg of KI as carrier in a total volume of 50µl at 70°C for 6 hrs. At the end of these experiments, the wires were washed thoroughly with double distilled water and dried under an infrared lamp. The approximate radioactivity on the wires was measured by an ionization chamber.

4.1.5. Internal electrolysis

The deposition of iodine-125 on silver wires was studied using the internal electrolysis method. In these experiments, 10 ml of electrolyte solution containing 0.01 M/l NaOH and both carrier-added and carrier-free iodine-125 was used. Concentration of carrier iodine-125 in the electrolyte was 1.6 x 10^{-4} M/l NaI. 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 1.1 GBq (30 mCi). From among different metals examined as counter electrodes, platinum was chosen for iodine deposition, as its standard potential is more positive than that of silver. The coating solution was placed in a Pt crucible that served as cathode. The uncut wires were immersed into the solution contacting the brim and bottom of the crucible. The deposition yield was determined by an indirect method. The residual non-adsorbed radioactivity in the plating solution and washings of the wires was measured using an ionization chamber. The plating assembly was placed in a glove box connected to a radioactive fume cupboard.

4.1.6. Electro-deposition

Anodic electro deposition of radioiodine (^{125}I) was carried out in a quartz bath size [1.2 cm (dia.), 2.5 cm (h)] with platinum cathode (1mm) in a cell. Thoroughly washed silver wires (3-4 numbers) were placed at the center of the cell as anode. Radio iodine solution 111-130 MBq (3-3.5 mCi)/30 µl was added to 0.01M Na_2SO_4, adjusted to pH 9 using 1mM ammonia and the total volume made up to 650µl. The deposited radioactivity was measured using a dosimeter. About ~84% of the activity can be deposited at a current of 20 mA for 25-30 min duration.

4.1.7. Physical adsorption on ceramic rod

For the ^{125}I brachytherapy seed, a carrying media consisting of Al_2O_3 and silver nano powder was developed. In this material, approximately 15 wt% of Ag is contained. The porosity of the material is approximately 10%. Each rod was contacted with a ^{125}I solution of 5 mCi (185 MBq) at 50µl volumes at alkaline pH (9~12) for 4 hours.

4.1.8. Vacuum sublimation

The method is based on thermal sublimation of free iodine in vacuum and subsequent chemosorption on substrates (core blanks), which can have various dimensions and shapes (plates, foils, microspheres, wires). Promising results were obtained, particularly with silver substrates. Maximum activity density on the substrate surface depends on surface preparation technique and vacuum level. Sorption of sublimated iodine on chemically cleaned silver in vacuum $1 \cdot 10^{-4}$ Torr gives 16-17 mCi (~ 600 MBq) of ^{125}I per square millimeter of the surface, providing achievable activity up to 70 mCi (2.59 GBq) in one standard brachytherapy seed. A fresh vacuum evaporated silver surface accommodates up to 23 mCi/mm^2 (about 3.7 GBq in one seed), but requires more complicated equipment. An iodine layer deposited by vacuum sublimation onto silver is thermally stable until ~500°C, and its leachability in water was found to be below 0.01% for 24 hours at room temperature. For batch production of ^{125}I cores for brachytherapy seeds, the iodine can be deposited onto long (1-2 m) silver wires (dia. 0.5 mm) rolled in the coil, or onto straight pieces of wire arranged in a compact assembly, which fits the vacuum sorption chamber, with subsequent cutting of the treated wires to 3 mm long bars.

4.1.9. Adsorption of iodine-125 on chlorinated silver bars

Hydrogen dioxide solution and hydrochloric acid were used for chlorination on silver bar surface. The optimal adsorption time and controlled pH value of silver bar iodination is over 15h and 5 respectively. The ^{125}I adsorption rate increases when the quantity of carrier and ^{125}I activity concentration increases. Experimental results showed that the uniformity adsorption of ^{125}I-Ag bar improves when the quantity of carrier increased. It is very important to control the adsorption temperature and the adsorption volume. Total self-shielding of ^{125}I seed source were close to 40-50%. Further work should confirm and improve these results.

4.1.10. Preparation of spherical type sources

Alumina micro spheres and metallic silver spheres were used as spherical sources.

Alumina micro spheres

A mixture of pre-cooled (5°C) solution of hexamethylenetetraamine and urea (3M) with aluminum nitrate solution was dispersed as droplets into hot oil to bring about the conversion to the solid gel sphere form. The spheres were separated from the oil and collected on a wire mesh, washed free of grease with carbon tetrachloride and then washed thoroughly with ammonia solution (2M) to remove the chemical impurities occluded inside the spheres. The spheres were dried in an air oven at 100°C and heat treated at 700°C for 5 h in a furnace. The uniform sized spheres were selected by passing through a 600 μm mesh. Five microspheres, previously conditioned with 1 mM sodium nitrate at pH 2-3, were immersed in ^{125}I (100 mCi/ml) followed by the addition of 0.1ml of freshly prepared chloramine-T solution (1 mg/ml in 1 mM sodium nitrate) in a quartz test tube. The tube was closed tightly and the microspheres were shaken using an auto shaker at ambient temperature (22°C). The spheres were washed twice in the 1 mM sodium nitrate to remove the non-adsorbed activity and dried at room temperature. Five microspheres, previously conditioned with 10^{-3}M sodium nitrate at pH 2–3, were immersed in ~30 μl of ^{125}I (100 mCi/ml) followed by the addition of 0.1ml of freshly prepared chloramine-T solution (1 mg/ml dissolved in 10^{-3} M sodium nitrate). The microspheres were allowed to stand in the radioactive iodine (IO^-_3) solution in a volume of ~130 μl in a quartz test tube closed tightly with stoppers and were shaken using an auto shaker for 4 h at room temperature (22°C ambient). The spheres were washed twice in the same medium (sodium nitrate solution) to remove the unadsorbed activity and dried at room temperature.

The percentage adsorption of iodine activity on the spheres was indirectly measured by counting the remainder of the adsorbing solution and the washings in a well-type scintillation counter set for ^{125}I activity, after sufficient dilution.

Metallic silver spheres (beads)

Silver beads of ~ 0.5 mm (dia.) were cleaned with 3M HCl followed by thorough washing with water. The coating of palladium on silver beads was carried out by treating the beads with $PdCl_2$ solution at ~100˚C for 15-20 minutes. The palladium-coated beads were washed and dried to constant weight. The beads were individually taken in small glass reaction tubes and treated with ~10µL of ^{125}I solution containing ~ 9.25 MBq (250 µCi) of ^{125}I at pH 9-10. Carrier iodide as KI was added to the individual reaction tubes to enhance the adsorption kinetics. The total reaction volume was maintained as 15µL and the reaction temperature was maintained at ~60-70ºC for 7 h. The adsorbed beads were washed with warm (~50ºC) distilled water. The activity in the bead after adsorption was measured by using a pre-calibrated ion chamber.

4.1.11. Source core encapsulation and welding techniques

Titanium is a unique material, as strong as steel but half its weight, with excellent corrosion resistance. The high strength, low weight and outstanding corrosion resistance possessed by titanium have led to its choice as the capsule material.

Although the tungsten inert gas (TIG) process is the most common method for welding titanium materials, this is not suited to this type of capsule, as it causes porosity and resulted in leakage of activity from the sealed source. To overcome this problem, Nd:YAG pulsed laser welding system was used by all the countries, except China, for the welding. Major advantages of laser beam welding are low welding stresses, low risk of distortion, creation of minimal heat affected zone with minimal ^{125}I contamination and the capability of welding varying mass that allows hermetic sealing of Ti capsules. The high quality of welds by electron beam was demonstrated by China.

4.1.12. Quality control testing

Radioactive source cores should be tested in order to examine:

- source uniformity testing by autoradiography or activity measurement with collimator equipment and
- leachability in water and saline solution.

Additionally, the prepared sealed source should be subjected to the following tests:

- immersion test (Leak Test),
- wipe tests (Surface Contamination Testing) and
- accurate measurement of activity.

The prototype seed sources should undergo the QC tests in accordance with ISO 2919. The class obtained is C 53242. This means that the seed-type sources maintained their leak tightness according to ISO 978 after testing against:

- temperature test, from - 40ºC (20 min.) up to +600ºC (1 h) and thermal shock 600°C to 20°C,
- external pressure test, from 25 kPa absolute up to 2 MPa absolute,
- impact – 50 g dropped from 1 meter,
- vibration – 3 x (10 min) 25 Hz to 80 Hz 1.5 mm amplitude peak to peak and 80 Hz to 2000 Hz at 196.2 m/s^2 (20g) peak acceleration amplitude and
- puncture – 0.001 kg puncture from 1 meter.

4.1.13. Calibration and dosimetry of sources

Accurate measurement of activity should be performed by a well chamber previously calibrated using primary/secondary standard.

The dosimetric parameters of the source can be derived in accordance with the recommendations of the AAPM Radiation Therapy Committee Task Group (TG)-43 (Dosimetry of Interstitial Brachytherapy Sources: Recommendations of the AAPM Radiation Therapy Committee Task Group No. 43. Medical Physics 22 (2), February 1995).

4.1.14. Conclusions

Two ways of manufacturing the source core using the electrochemical method were considered: a) coating longer Ag wires with iodine-125 and subsequently cutting into appropriate pieces (Poland) and b) coating with radionuclide pre-cut bars 3 mm long (India).

Physico-chemical adsorption of ^{125}I activity on the $PdCl_2$ treated silver was investigated and adopted by India, Peru and Iran. In this method, quantitative adsorption of ^{125}I was possible. The mild experimental condition of adsorption at neutral to alkaline pH facilitates the safe handling of high amounts of radioactivity for the preparation of ^{125}I sources without the release of air activity. It was possible to prepare sources up to 4 mCi (148 MBq). Such a method assures a very low leaching level (0.01%).

The method of adsorption of iodine-125 on chlorinated silver bars (China) was applied for the preparation of sources. About 90 % of ^{125}I can be adsorbed using this method. Sources up to activities of 1 mCi (37 MBq) could be prepared with low leachability.

A sublimation method was investigated using iodine-131 as tracer (Kazakhstan). The method is based on thermal sublimation of free iodine in vacuum and subsequent chemosorption on substrates (core blanks) of various shapes and sizes. Various chemical and physical parameters were standardized to obtain quantitative uptake of iodine on the substrate. The advantage of this technique is that activities up to 70 mCi (2.59 GBq) could be deposited on the source matrix.

The absorption of iodine-125 in ceramic rod treated with silver nano particles was investigated in Korea. Preliminary studies show that a ceramic rod treated with silver nano particles has a good loading capacity for iodine. Quantitative adsorption of activity on the rod is possible and sources up to 5 mCi (185 MBq) have been prepared. The investigation is still in progress and more investigations are needed to optimize various parameters.

An internal electrolysis technique has been developed and adopted in Poland and is characterized by its simplicity and effectiveness. By this method it is possible to obtain four coated silver bars, each 12 mm long and 0.5 mm dia., containing 140 mCi (5.18 GBq) of ^{125}I activity, each in a single procedure, although this number can be increased depending upon demand. A cutting machine has been designed and fabricated for automatic cutting of silver wires coated with ^{125}I into 3 mm long pieces and positioning of the source cores inside titanium capsules. The results confirmed that contamination of the source capsules can be minimized employing the above technique.

Similarly, anodic electro deposition of radioiodine (^{125}I) has been tried in India using silver electrodes 3 mm long and 0.5 mm in diameter, and various experimental parameters were optimized to obtain quantitative adsorption. However, the leachability of these sources was much higher than that of palladium coated sources. Efforts are being made to reduce the leaching by having an organic coating. This work is still in progress.

As far as preparation of spherical type sources are concerned, two types of sources such as alumina microspheres and metallic silver spheres have been investigated (India). It was found that the percentage leachability of radioactivity from the alumina spheres was slightly more than desirable. More investigations are needed to overcome this problem. The physicochemical adsorption of Pd-silver microspheres is easy and ^{125}I-beads could be prepared in a non-leachable form.

As the source design of small radioactive sources plays a vital role in the dosimetric aspects, the spherical ^{125}I-beads on account of relatively higher dose rate delivery may reduce the total treatment time and potentially replace the presently used rod type ^{125}I-sources in eye and prostate brachytherapy.

To summarize, the preparation of ^{125}I sources, based on various techniques that produce high fixation yields with good reproducibility, amendable for large scale production, was achieved through the CRP. Knowledge for laser welding of the sources encapsulated in titanium capsules was also gained. These sources can be used for the treatment of ocular and prostate cancers.

4.1.15. Application and uses of iodine-125 seed-type sources

^{125}I sources are used in the radiotherapy of brain, neck, lung, pancreas and prostate cancers, as well as intraocular tumours (choroidal melanomas and retinoblastomas). They can be used in two different ways – permanent and temporary implants. However, the temporary implants are used only as booster doses using high amounts of radioactivity. ^{125}I seeds are indicated for the treatment of tumours that have the following characteristics: localized, slow growth rate and low to moderate ratio sensivity. They are also indicated for the treatment of recurrent tumours and residual tumours following a course of external radiation therapy. Required activity of ^{125}I seed used in ophthalmic applicators is, on average, 20 mCi (740 MBq) and in other application most often 4-5 mCi (148-185 MBq).

Clinical trials of these sources were carried out in one of the leading eye care hospitals in India in which 5 patients were treated using locally prepared seeds. Out of these five cases, there were two cases of retinoblastoma and three cases of malignant melanoma. Although the number is very low to draw definitive conclusions for the efficacy of the treatment, it is highly encouraging to know that brachytherapy using ^{125}I sources has been effective in a certain category of patients and enucleation of the eye could be prevented. It is also noteworthy that external beam radiotherapy often fails in the treatment of ocular tumours, which respond well to the brachytherapy treatment.

4.2. Palladium-103

4.2.1. Primary radioisotope production

^{103}Pd is commonly produced by cyclotron irradiation of rhodium targets with accelerated protons. This approach allows production of carrier-free palladium-103 having near theoretical value of specific activity of 75000 Ci/g (~2.8 10^6 GBq/g). Nevertheless, carrier (metal palladium) is commonly added to stabilize its behaviour at isolation/purification from radioactive impurities and at seed core production, as well. In such a case the ^{103}Pd specific activity decreases, but it should be at least 5 Ci/g (185 GBq/g) for successful source production. The disadvantage of cyclotron production of ^{103}Pd is the relatively high price of this radioisotope.

An alternative approach to ^{103}Pd production is reactor irradiation of isotopically enriched ^{102}Pd. The low natural isotopic abundance of ^{102}Pd (1,02%) requires the use of moderate to highly enriched ^{102}Pd target material of no less than 50%. Moreover, the low cross-section of ^{102}Pd(n,γ)^{103}Pd nuclear reaction (thermal cross-section is 3,4 barn, resonance integral 10 barn) requires also the use of high-flux nuclear reactors, like the HFIR (ORNL, Oak-Ridge, USA) or SM (RIAR, Dimitrovgrad, Russia), to produce high-specific activity ^{103}Pd.

Unfortunately, the enrichment of palladium with ^{102}Pd is rarely exceeds 50-80%, i.e. this material contains other palladium isotopes, like ^{108}Pd and ^{110}Pd. These isotopes can be activated by neutron irradiation, for example ^{108}Pd produces ^{109}Pd ($T_{1/2}$ = 14 h) that cannot be separated from ^{103}Pd by chemical means.

In another nuclear reaction ^{110}Pd produces ^{111}Pd ($T_{1/2}$ = 23 min) that decays to ^{111m}Ag ($T_{1/2}$ = 7.5 d), thus radiochemical procedure should be applied to purify ^{103}Pd from the radioactive silver impurity.

Specific activity of the reactor-produced ^{103}Pd depends on the target material enrichment, neutron flux density and irradiation duration. At the Oak-Ridge National Laboratory, USA, ^{103}Pd was produced from ^{102}Pd with 79.2% enrichment. The target was irradiated in a neutron flux of 2 x 6.10^{15} $cm^{-2}s^{-1}$ for 2 reactor campaigns (21 days each with 3 days intermediate shut-down). Irradiation duration was limited by activity of radioactive ^{111}Ag produced from ^{110}Pd. Under these conditions the ^{103}Pd specific activity was 345 Ci/g (~1.3 10^4 GBq/g).

For the SM reactor operated by RIAR the experimentally determined specific activity value was 495 Ci/g (~ 1.7 10^4 GBq/g) after an irradiation period of 19.2 effective. days. Thermal neutron flux density inside the target volume was 1 x 8×10^{15} $s^{-1}cm^{-2}$, and that of epithermal neutrons was 1 x 65×10^{15} $s^{-1}cm^{-2}$. The target isotopic composition was as follows: ^{102}Pd - 89.0%, ^{104}Pd – 3.6%, ^{105}Pd – 4.0%, ^{106}Pd – 2.45%, ^{108}Pd – 0.75% and ^{110}Pd - < 0.2%.

Post irradiation treatment included mechanical cutting of the titanium capsule and destroying the quartz one. Palladium metal was dissolved in 10 ml of the HCl and HNO_3 (1:3) mixture.

High enrichment of target material provides low activity of ^{101}Pd and ^{111}Ag impurities, so that no radiochemical purification of ^{103}Pd is required. Low enriched material after irradiation can be purified (mainly from ^{111m}Ag) by well-established ion-exchange procedures.

Moderate flux reactors with neutron flux density of 1-2 x 10^{14} $cm^{-2}s^{-1}$ can also be used for ^{103}Pd production, as that should provide ^{103}Pd specific activities much higher than the required minimum of 5 Ci/g (185 GBq) for brachytherapy seed production.

4.2.2. Technical requirements of the source

Requirements of ^{103}Pd sources are close to those applied to ^{125}I sources, as medical applications of sources are quite similar. They are as follows:

- source activity should be 0,5 – 5 mCi (18.5 – 185 MBq) with less then $\pm$10% deviations,
- source dimensions should be 0.8 mm dia and 5 mm long,
- the source capsule should be made of titanium,
- source core dimensions should be 0.3 mm dia and 3 mm in length,
- distribution of ^{103}Pd along the source should be uniform and
- the source should be leak tight.

The process of preparing sources in large batch sizes should be reproducible and feasible in a minimum amount of time.

4.2.3. Source core preparation

Electroplating of palladium on various wire-type supports made from palladium or nickel metal can be used for production of palladium-103 seed core.

Plating is carried out using regular electrochemical cells, arranged for single- or multiple cathode placements. The cell body can be made of plexiglass, the anode can be made of rhodium or iridium foil. Twisting of the cathode should be prevented by special means, like fixing it in a centering hole or by using a specially designed cathode holder. Forced cell cooling should be provided during electrochemical plating.

Cathodes are made of nickel wire of 0.3 mm diameter and 30-35 mm in length. Before palladium electroplating the support surface is treated (activated) with electrolyte of the following composition:

20 ml of H_2O 2 ml of HNO_3 (conc.) and 2 ml HCl (conc.).

The nickel wire is effectively activated for 30 sec at 50°C or 1-2 min at ambient temperature. After that, the wire is flushed with water and ethanol.

After pre-treatment the support is fixed in the cathode holder. The cell is filled up with the electrolyte, the cathode is placed inside the cell and plating proceeds using the selected mode (current, voltage, time). After the plating is completed, the power supply is switched off, the cathode is taken out of the cell and out of the cathode holder, rinsed with water, ethanol and dried under vacuum for 10 min. The convenient DC power source is used for power supply.

Layers with good adhesion to the wire support and homogeneous distribution of palladium can be produced by using an electrolyte of the following composition: H_2PdCl_4 -3,5 g/L, $(NH_4)_2HPO_4$ - 20 g/L, $Na_2HPO_4 \cdot 12\ H_2O$ -100 g/L, NH_4Cl -25 g/L, NH_4OH -adjusted to pH 7. The electrolyte is prepared by mixing water solutions of its components in the following sequence: $H_2PdCl_4 \rightarrow (NH_4)_2HPO_4 \rightarrow Na_2HPO_4 \cdot 12\ H_2O \rightarrow NH_4Cl$. The solution is diluted with water up to the desired volume and NH_4OH is added to adjust pH to 7. Palladium is plated at a current density of 0.25 A/dm^2 at 50°C. The yield of palladium is 90-95%.

For a wire length of 35 mm the uniformity of ^{103}Pd deposition is better than 95% for the central section of the wire and better than 90% for its ends. The wire can be cut into pieces 3 mm long, and they can be used for seed production.

4.2.4. Source core encapsulation and welding techniques

The techniques for source encapsulation as well as capsule welding used for ^{103}Pd seeds production are similar to those applied for the production of ^{125}I seeds, described above.

4.2.5. Quality control testing

The prototype seed sources should undergo the QC test in accordance with ISO 2919. The class obtained is C53242. This means that the seed-type sources maintained leak tightness according to ISO 978 after testing against:

- temperature test from - 40º C (20 min.) up to +600º C (1 h) and thermal shock 600°C to 20°C,
- external pressure test from 25 kPa absolute up to 2 MPa absolute,
- impact – 50 g dropped from 1 meter,
- vibration – 3 x (10 min) 25 Hz to 80 Hz 1.5 mm amplitude peak to peak and 80 Hz to 2000 Hz at 196.2 m/s^2 (20g) peak acceleration amplitude,
- puncture – 0.001 kg puncture from 1 meter.

Source cores should be tested for the following examination:

- source uniformity testing by autoradiography or activity measurement with collimator equipment,
- leachability in water and saline solution.

Additionally, the prepared sealed sources should be subjected to the following tests:

- immersion test (Leak Test),
- wipe tests (Surface Contamination Testing) and
- accurate measurement of activity.

The source cores should be tested for uniformity by autoradiography or activity measurement with collimating equipment. The sealed source should be subjected to the immersion (leak) test, wipe test (surface contamination test) and accurate measurement of source activity.

4.2.6. Calibration and dosimetry of sources

Accurate measurements of source activity should be performed by suitable counter previously calibrated using primary/secondary standards.

It should be noted that up to 35% of ^{103}Pd radiation is absorbed by the titanium capsule and the quantity of radioisotope plated on the support wire should consider these "losses".

4.2.7. Conclusions

The production technology of ^{103}Pd by nuclear reactor irradiation of isotopically enriched ^{102}Pd has been developed and the radionuclide can be produced in high specific activities and can also be supplied to interested parties for sealed sources production. A technique for seed core preparation has been developed. It can be applied for source production technology by anyone who has access to ^{103}Pd produced either by cyclotron or reactor irradiation.

4.3. Iridium-192

4.3.1. ^{192}Ir HDR source for brachytherapy

The main advantage of these sources is relatively low average energy of γ-radiation (0.412 MeV) that would make local protection of critical organs and tissues easy. That is why the ^{192}Ir sources for brachytherapy are the most interesting for medical purposes. There are a variety of specialized computer controlled remote after loading systems like a GM 12i type 444 GBq (12 Ci). This system uses a high activity point source of ^{192}Ir. Iridium in the form of wire is used for the treatment of tumours in the breast, tongue and other tumours.

4.3.2. Production of primary radioisotope

^{192}Ir is produced by (n,γ) reaction on iridium in a nuclear reactor. ^{192}Ir decays with a half-life of 74.2 days to stable ^{192}Pt and emits beta particles of maximum energy of 0.672 MeV (46%) and gamma rays of energies of 0.604, 0.468 and 0.308 MeV. When preparing ^{192}Ir γ ray sources, the influence of neutron self-shielding during irradiation caused by its high neutron capture cross section is a problem in obtaining intensive sources with a high specific radioactivity. The active core is a set of disks with a diameter of 0,6 x 3,5 mm wire length, or a diameter of 0,5 x 0,5 mm length of the pellet (iridium metal, 99.9% pure). This iridium target is irradiated in a high flux reactor, providing more than 2x 10^{14} n.cm^{-2}.sec^{-1} neutron flux density, because specific activity of the irradiated target should be more than 16.8-18.5 TBq/g (450-500 Ci/g). Activity of the ^{192}Ir HDR assembly should be more than 370 GBq (10 Ci) for effective therapy.

4.3.3. Source preparation

The sealed source capsule (diameter 1,1 mm, length 4,6 mm) and plug for brachytherapy is made of stainless steel and is connected to flexible stainless steel wire (+ 2100 mm in length) with end plug. As usual, non-active parts are welded at ordinary conditions by means of laser welding.

4.3.4. Source encapsulation and welding

Several investigations into welding methods for encapsulation of the sources were provided.

In the Republic of Korea, the capsule is made from two parts and was welded from the sides with a laser welding machine. In Hungary they weld the capsule to the extension wire by laser welding, and they weld the capsule with the plug on top by microplasma welding. In Belarus they weld the capsule with the plug on top by laser welding.

For welding the capsule, the following parameters of the laser welding system are recommended:

- output of the laser machine: more than 30 W,
- diameter of the laser beam: 120-250 μm,
- depth of the welded joint: more than 200 μm,
- inert gas: He or Ar,
- applied voltage: 200- 250 V and
- frequency: 10-28 Hz.

The participant from Iran designed and developed a brachytherapy source assembly for after loading. The diameter of the capsule is 4.6 mm and its length is 6.5 mm, allowing the use of argon arc welding. For capsules with smaller dimensions the laser welding method is recommended.

The mechanical system for assembly fabrication should have a stable base for supporting the parts of the system, because it should be insulated against vibration. The target to be welded should be positioned and rotated around the longitudinal axis with a special mechanical system. A PC control system is needed for operating the machine and controlling and adjusting the following parameters:

- control of required motion,
- control of target rotation,
- control of laser-impulse,
- control of welding parameters and
- control of automatic welding process.

Each of the sealed sources should be provided with a written “Source Certificate”. This certificate contains the most important significant data of the sealed source concerning the International Requirements.

4.3.5. Quality control methods

Radioactive sources are manufactured in accordance with strict quality control methods, details of which can be obtained on request. Stringent tests for leakage are an essential feature of radioactive source production. The methods adopted depend on the design and intended application of the source, and also on statutory requirements. Where necessary, tests can be specially modified to meet particular requirements. The standard methods such as ISO 1677 “Sealed Radioactive Sources-General”, ISO 9978-1992(E) “Radiation protection-Sealed radioactive sources-Leakage test methods” and ISO 2919: 1999(E) “Sealed Radioactive Sources-Classification” should be used for quality control tests of radiation sources. According to the ISO 2919: 1999(E), ^{192}Ir brachytherapy sources used to correspond with the ISO C53211 class of strength.

A test report is supplied with each source or batch of sources. When changing the design and technology for manufacturing, the source must be tested according to ISO 2919: 1999(E). According to the demands of ISO 2919: 1999(E) measuring is also a part of the QC program. Participants from Belarus and Iran use similar methods of measuring ^{192}Ir source dose rate by means of the Farmer chamber 300001 PTW calibrated for 250 kV X ray and ^{60}Co.

For loading the assembly into medical equipment, an applicable test should be carried out as follows:

- loading/unloading test to verify structural endurance of the source assembly,
- exposure test to observe accuracy of source positioning,
- experiments for the source movement and the accuracy of positioning in various brachytherapy applicators,
- experiments to evaluate a source-driving obstacle which occurred by the curvature of the source-moving pathway and
- measurement of activity and anisotropy of the source assembly.

4.3.6. Conclusions

The participating group from Belarus has already started the batch manufacture of brachytherapy sources based on the technology described in this project. They have already received the Special Form Radioactive Material Approval Certificate. A few brachytherapy sources have already been installed and used in afterloading machines.

4.4. ^{192}Ir industrial sources

^{192}Ir is one of the most widely used sealed sources for NDT applications. Taking into account the relatively short half-life of the isotope (74 days), it is very essential to organize the manufacture of sources as close as possible to the end user to avoid logistic problems.

4.4.1. Primary isotope production

^{192}Ir is produced by (n,γ) reaction on iridium in a nuclear reactor. ^{192}Ir decays with a half-life of 74.2 days to stable ^{192}Pt and emits beta particles of maximum energy of 0.672 MeV (46%) and gamma rays of energies of 0.604, 0.468 and 0.308 MeV. When preparing ^{192}Ir γ ray source, the influence of neutron self-shielding during irradiation caused by its high neutron capture cross section is a problem in obtaining intensive source with a high specific activity. The active core is a set of disks with diameter of 0.5-3.5 mm and height of 0.2-0.5 mm. These iridium targets are irradiated in a moderate to high flux reactor, providing more than 2×10^{14} n.cm^{-2}.sec^{-1} neutron flux density, because specific activity of the irradiated target should be about 250-400 Ci/g. Activity of the ^{192}Ir source should be about 20-150 Ci for effective NDT.

4.4.2. Source preparation

The sealed industrial sources ^{192}Ir assembly consists of a stainless steel capsule with or without holder, active core as a set of irradiated disks and stainless steel plug. The participant from Belarus uses the design of capsule without a holder.

The procedure of source preparation consists of three steps:

- the necessary number of irradiated disks is put inside the capsule by means of a vacuum pipe,
- the plug is installed on top of the capsule by means of a special pressure device and
- the assembly is moved to another hot cell, where the welding machine is placed.

4.4.3. Source encapsulation

Sealing is carried out using argon - arc circular edge weld. The preferable welding regimes have been found to be:

- current strength - 13-14 A,
- rotation speed - 3.0-3.3 rpm,
- overhang of capsule from tong - 0.5 mm,
- arc length - 0.8-1.2 mm and
- argon consumption - 4-10 l/min.

4.4.4. Quality control methods

Radioactive sources are manufactured in accordance with strict quality control methods, details of which can be obtained on request. Stringent tests for leakage are an essential feature of radioactive source production. The methods adopted depend on the design and intended application of the source, and also on statutory requirements. Where necessary, tests can be specially modified to meet particular requirements. The standard methods such as ISO 1677 "Sealed Radioactive Sources-General", ISO 9978-1992(E) "Radiation protection-Sealed radioactive sources-Leakage test methods" and ISO 2919: 1999(E) "Sealed Radioactive Sources-Classification" should be used for quality control tests of radiation sources. According to the ISO 2919: 1999(E) ^{192}Ir industrial sources use to correspond the ISO C43515 class of strength.

A test report is supplied with each source or batch of sources. When changing the design and the technology for manufacturing, the source must be tested according to ISO 2919: 1999(E). According to the demands of ISO 2919: 1999(E) measuring is also a part of the QC program. The participant from Belarus adjusted the method for measuring brachytherapy sources to industrial ones.

4.4.5. Conclusions

The participating group from Belarus has already started the batch manufacturing of industrial ^{192}Ir sources based on the technology described in this project. They have already received the Special Form Radioactive Material Approval Certificate. A few dozen sources have already been installed and used in the NDT projectors.

4.5. Ytterbium-169

4.5.1. Primary isotope production

It is advisable that the ^{169}Yb source has approximately 185 ~ 370 GBq (5 ~ 10 Ci) activities as a point source to apply to a radiographic industrial NDT application. The carrier free ^{169}Yb can be produced by cyclotron irradiation of ^{169}Tm via ^{169}Tm (d,2n) ^{169}Yb reaction. However, this method has limitations due to an unstable supply and limited production quantity. Hence, neutron irradiation is more suitable for a stable supply and mass production of ^{169}Yb.

The production scheme of ^{169}Yb by neutron irradiation uses the ^{168}Yb (n,γ) ^{169}Yb reaction. Since a natural ytterbium target contains ^{168}Yb at only 0.13%, an enriched ytterbium target is required in order to attain a proper specific activity. Irradiation of the target should be accomplished in the conditions providing ^{169}Yb specific activity 1200 – 2400 Ci/g. Moderate- and high-flux reactors can be used for irradiation. Examples are the HANARO research reactor (Republic of Korea), the SM research reactor (Russia) etc.

4.5.2. Technical requirements of the source

- Source design (outer capsule and holder) and dimensions should make available the replacement of ^{192}Ir sources with ^{169}Yb sources for use in conventional NDT projectors.
- Source design should also provide its safety, so double encapsulation is preferred. Materials used for source encapsulation should have low Z, preferably – aluminium or titanium, to ensure low absorption of radiation by capsules.

- Source core dimensions should be as small as possible, to ensure high resolution of the NTD device, preferably 1x1 mm or less. Besides, the source core material should have a low leaching ability.

4.5.3. Preparation of Yb core, target and reactor irradiation

^{169}Yb source core developed at Korea Atomic Energy Research Institute (KAERI) is a cylindrical pellet. To fabricate the cylindrical Yb_2O_3 pellet with a diameter of 1mm and length of 1mm, compacting equipment and a die have been developed. The procedure of pellet production includes pressing and sintering. The density and hardness of the pellets increases as the compaction force is raised, and the best hardness could be achieved at a 100 kg compaction and an 1150^{o}C sintering for three hours. At these conditions the pellet density is approximately 6.0 mg/mm^3 and irradiation in the HANARO reactor provides a pellet activity of approximately 7 Ci (259 GBq).

To produce the irradiation target, the pellet is encapsulated in the aluminum capsule (outer dimensions after welding are 4.274 mm length, 1.875 mm diameter), which is irradiated in the HANARO reactor. The welding of the aluminum capsule is performed by using an Nd-YAG pulse laser. Welding conditions are as follows: beam speed 80 mm/min, pulse frequency 20 Hz, pulse width 2 msec and shielding atmosphere – Ar gas, 15 l/min. The targets are irradiated in a thermal neutron flux of 2.13×10^{14} n.cm^{-2} • sec^{-1} during 23 days.

4.5.4. Design of source assembly

Two different capsule assemblies are designed for ^{169}Yb sources. In both cases, ^{169}Yb sources are enclosed in ^{192}Ir source capsules in order to use the NDT projectors for the ^{192}Ir source assembly in the market. Connector, pig tail, and capsule are the same as that of the ^{192}Ir source assembly, except that the ^{169}Yb source is installed with a left-handed screw fastening in the capsule and the pig tail is 3.8 mm longer than that of ^{192}Ir source assembly. The second assembly is designed to reduce the radiation absorption by the source capsule in which the ^{169}Yb source is installed as it is welded inside the aluminum capsule.

4.5.5. Activity calibration methods

A simple method to determine source activity is a relative measurement of dose rate using a certified standard source as reference, when it is available.

For measurement of absolute activity of radioisotope contained in the sealed source and determination of calibration factors, two basic methods are applied:

- Destructive method, which implies a complete digestion of the sealed source and measurement of activity by liquid scintillation counter. This requires designing a special chemical procedure in every specific case for quantitative transfer of radioactivity into liquid scintillator.
- Precise microcalorimetry, which requires complicated equipment but may be done without destruction of the source.

As each of these methods still has its own uncertainties, the preferred general approach is to use both of them, as well as both measurement and Monte Carlo calculations of the dose or dose rate, and compare the results.

4.5.6. Conclusions

A technology for ytterbium oxide pellet production has been developed by the Korean participant. A laser welding technique was developed for sealing aluminium and titanium capsules with Yb_2O_3 pellets, as well as for source production. The Korean participant has developed sources

design compatible with existing NDT projectors. These sources, when used instead of ^{192}Ir analogue, provide a higher quality radiography pattern.

5. POSSIBLE AREAS OF FURTHER RESEARCH

The developmental research efforts in producing miniature, compact high intensity sealed sources for brachytherapy, and enlarging the list of potential radioisotopes with special characteristics for specific applications, will continue in the future for medical as well as for industrial applications. These investigations will include improving existing source core preparation techniques to increase the levels of total radioactivity deposited, improved and simplify encapsulation and welding techniques and make them more amendable for automation and, to standardize and simplify quality control procedures and techniques.

The CRP participants have identified areas for future development that the Agency may consider for further promotion in its Member States. In particular:

- Development of improved technology for manufacturing ^{75}Se sealed radioactive sources for NDT applications as potential sources for higher imagine quality, compactness of the projector and less radiation exposure to the operator.
- Further development in the laser welding method for better quality of welding and increasing the levels of automation for larger scale production of ^{192}Ir industrial sources.
- Investigation in the production methods of X ray sources using ^{125}I and ^{153}Gd for possible application in low energy X ray NDT (50-150 mCi ~ 1.85-5.55 GBq)

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REPORTS BY PARTICIPANTS IN THE COORDINATED RESEARCH PROJECT

THE DEVELOPMENT AND MANUFACTURING OF MINIATURE SEALED SOURCES FOR BRACHYTHERAPY AND INDUSTRIAL APPLICATIONS

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Abstract

The aim of present work was to develop the methods, equipment and technology for assembling and sealing the ^{192}Ir sources, as well as to develop the principles of Quality Control (QC) procedures for brachytherapy and industrial sources. A few sets of welding experiments were carried out. The experiments were performed on stainless steel dummy source assemblies by means of laser welding machine in the hot cell. For sealing the industrial sources the argon arc welding was used. The analysis of the welding results was made by examinations of the metallography microsections. Pilot batch of dummy sources was produced and investigated. The optimal regime of welding was found. The QC programme was developed for brachytherapy and industrial sources. The QC programme consists of external examination, temperature tests, mechanical tests and measuring method. Finally the standard catalogue of sources was established and certificates for Special Form Radioactive Material were obtained. The development of the QC programme was in accordance with the requirements of the ISO 2919:1999E and GOST 25926-90. The State Committee for Metrology and Standardization and the Ministry of Health approved the part of programme for medical sources.

1. INTRODUCTION

The sealed HDR ^{192}Ir sources are commonly used for teletherapy and brachytherapy of malignant diseases in devices such as Microselectron and VariSource that are prevalent in oncological centres of Republic of Belarus [1, 2]. The main advantage of these sources is its relatively low average energy of γ-radiation (0.412 Mev) that would make easy a local protection of essential organs and tissues [3, 4]. That is why the ^{192}Ir sources for brachytherapy are of the most interest for medical purposes, now. The sealed ^{192}Ir sources are commonly used for non-destructive testing, as well. Those sources are widely adopted in field defectoscopes like GAMMARID that are prevalent in Belarus, now.

One of the main tasks of source manufacturing is the procedure for improving source properties such as decreasing radioactive leakage in case of unforeseen source damage. The development of the sealed sources encapsulating methods was the great part of the work in the scope of present CRP project. Development of quality control methodologies is another area of investigation. The adjustment of methods for assembling and sealing the sources was carried out by means dummy sources in hot cell with remote manipulator. The power unit and monitor of welding system were located in operator zone, the welding system, unit for micro-transference and rotating unit for dummy source was located inside the hot cell in “active “zone. The essential task of this work is to find out the preferable welding regimes and to adjust the mechanical and optical parts of welding systems.

2. MATERIALS

The irradiated ^{192}Ir was provided by RIAR (Dimitrovgrad, Russia). Capsules, plugs and wires were provided by Gamma-Service Recycling GmbH (Leipzig, Germany) and RIAR (Dimitrovgrad, Russia) for brachytherapy sources. Capsules and plugs for industrial sources were provided by Minsk Watch Plant (Minsk, Belarus).

3. METHODS

3.1. Sealing the ^{192}Ir sources for brachytherapy

The sealed source assembly for brachytherapy is the stainless steel capsule (1) (cylinder, diameter 1,1 mm, length – 4,6 mm) with plug (6) welded to flexible steel wire (4) (Fig. 1).

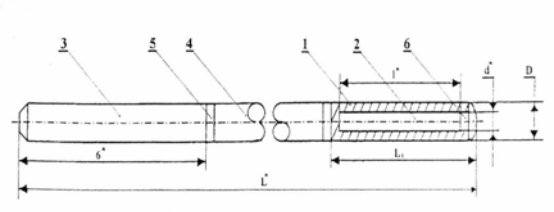

FIG. 1. Design of brachytherapy source assembly

The tail (3) was welded from the other side of the wire. It was developed the loading unit for remote load of the irradiated material imitator (2) (diameter 0,5 mm, length – 3 mm) into the capsule. The plug was mounted on the top of the capsule with help of this unit before welding. We used two different bushes, one for imitator loading and another for plug mounting. The welding marker was aimed on welding juncture by means of micro-transference unit of laser welding system. The design of loading and rotating unit did not allow blowing the weld zone by argon in laser emission direction [5]. That is why, first, the capsule was blown by argon athwart to the laser beam direction. The diameter of nozzle was 3 mm, the distance between nozzle and capsule was 5 mm. The rotating speed of capsule was 2 R.P.M. The distance between the welded object and objective was made in such way to attain the diameter of the welding spot 160-180 μm. Welding regimes are shown in the Table I.

TABLE I. REGIMES OF WELDING

No	Discharge time, ms	Pump voltage, V	The depth of weld, μm
1	3	240	190
2	3	260	250
3	3	280	400

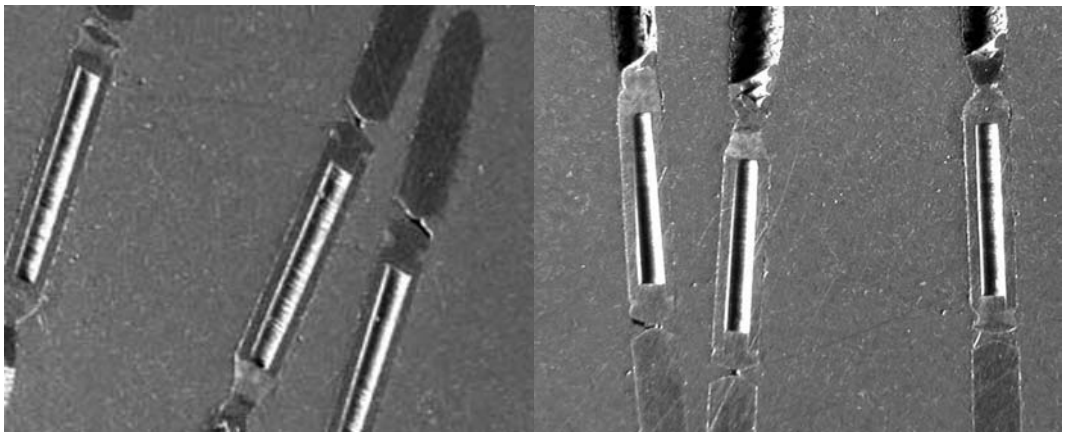

FIG. 2. Microsections of dummy sources

Investigating the microsection (Fig. 2) of welds we observed that:

- The surface of weld was oxidized
- There was metal blow-out at high voltage excitation
- The last pulse of laser emission was of very high energy and knocked out on welded surface deep lune
- The weld was not slick.

Placing the special mechanic lock on laser emission direction eliminated the effect of knocking out the lunes. Optical system was refocused for increasing the weld depth. The focus of the laser beam was rendered inside the metal when the image on the monitor became nice. The width of weld was increased up to 300 μm.

For decreasing the weld oxidation we did the following:

- The diameter of nozzle was made smaller – 1.5 mm.
- The distance between nozzle and welding surface was shortened for 2 mm.
- The angle between laser beam and argon blowing direction was set up 30^o (instead 90^o).

Welding regimes are shown in the table II. After mentioned above adjustments the weld looked smooth, bright and without oxidation (Fig.3).

TABLE II. REGIMES OF WELDING

No	Discharge time, ms	Pump voltage, V	The depth of weld, μm
1	3	250	200
2	3	260	280
3	3	265	320
4	3	270	380
5	3	280	through

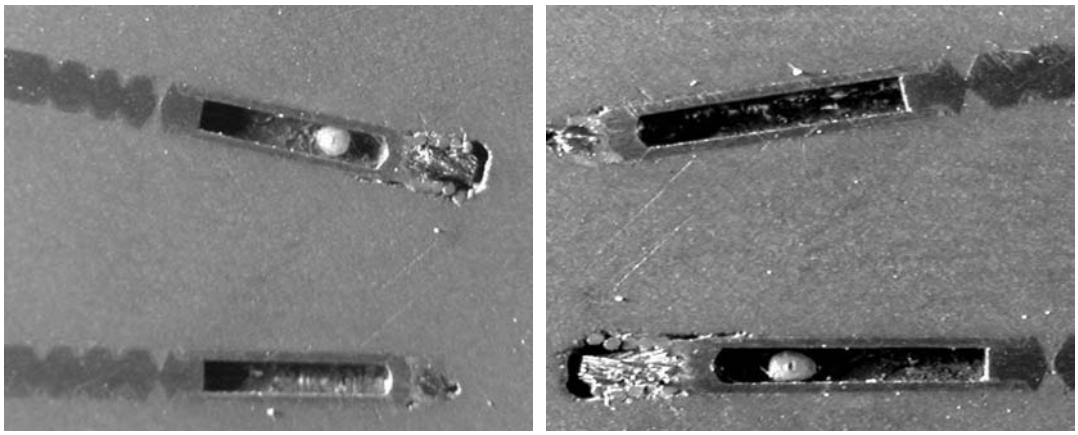

FIG. 3. Microsection of dummy sources.

There was produced a pilot batch of dummy sources. Dummy sources were tested by the commission of experts [2]. The following conclusions were made:

- Preparation for welding should include following operations:

- boiling in 10% solution Trilon-B, time – 10 min;
- boiling in distilled water, t=10 min;
- washing in alcohol;
- temperature drying 110-120 ^{0}C, t=10 min.

- The preferable welding regime should be following:

Pump voltage should be 240-260 V, discharge time should be 3-4 ms, and argon consumption should be 2-10 l/min.

- Results of welding

Visual inspection should be applied. Figure 4 shows microsection of a branchytherapy source. There are no craters, pores, cracks and other essential welding defects.

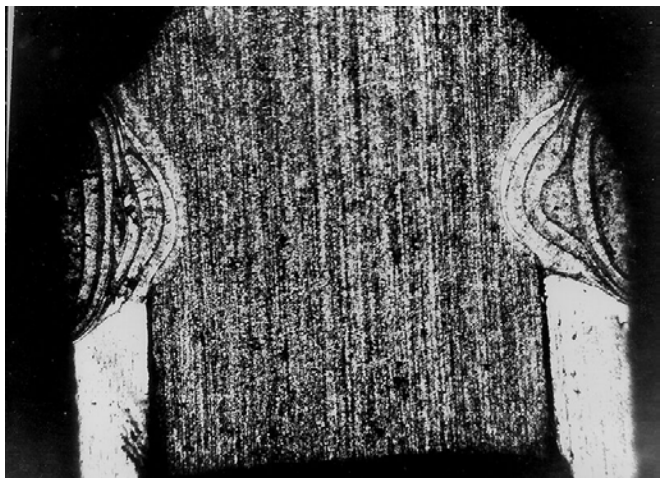

FIG. 4. Microsection of dummy brachytherapy source.

Dimension inspection was performed:

- diameter of capsules in welded points is $1.0^{+0.1}$ mm,
- length of capsule after welding is $5^{+0.1\ -0.04}$ mm

The inspection of dimensions shows that source dimensions correspond with specification (Table III).

TABLE III. SPECIFICATIONS FOR BRACHYTHERAPY SOURCES.

Source model	Overall dimensions, mm						Dose rate of air Kerma at 1 m distance mGy/h	Equivalent activity Bq (Ci)
	source		source with holder		active core			
	D1	L1	D	L*	D*	l*		
GI192M11.410							4.16	$3.7\ 10^{10}$ (1)
GI192M11.111							12.24	$1.1\ 10^{11}$ (3)
GI192M11.211							19.58	$1.8\ 10^{11}$ (5)
GI192M11.311							31.82	$2.8\ 10^{11}$ (7.5)
GI192M11.411	$1.0^{+0,1}$	$5^{+0,1\ -0,5}$	$1.0^{+0,4}$	2100	0.6	3.5	41.6	$3.7\ 10^{11}$ (10)
GI192M11.311-1	$1.1^{+0,1}$						40.75	$3.4\ 10^{11}$ (9,2)
GI192M11.311-2	$1.0^{+0,1}$	$5^{+0,1-0,5}$		1500	0.6	3.5	40.75	$3.4\ 10^{11}$ (2)
GI192M12.410							4.16	$3.7\ 10^{10}$ (1)
GI192M12.610							6.24	$5.6\ 10^{10}$ (1,5)
GI192M12.111							12.24	$1.1\ 10^{11}$ (3)
GI192M12.211							19.58	$1.8\ 10^{11}$ (5)
GI192M12.311							31.82	$2.8\ 10^{11}$ (7,5)
GI192M12.411	$1.0^{+0,1}$	$5^{+0,1-0,5}$	$1.0^{+0,1}$	1000	0.6	3.5	41.6	$3.7\ 10^{11}$ (10)

3.2. Sealing the industrial ^{192}Ir sources.

^{192}Ir industrial sources (Fig.5) are sealed radioactive sources without holder. The capsule and plug were made from chromo-nickel stainless steel 12X18H10T (quality approved by manufacture's certificate). There were prepared two sets of non-active accessories for sealing the radiography dummy sources type GI192M54 and GI192M55 (Table IV, Figure 5).

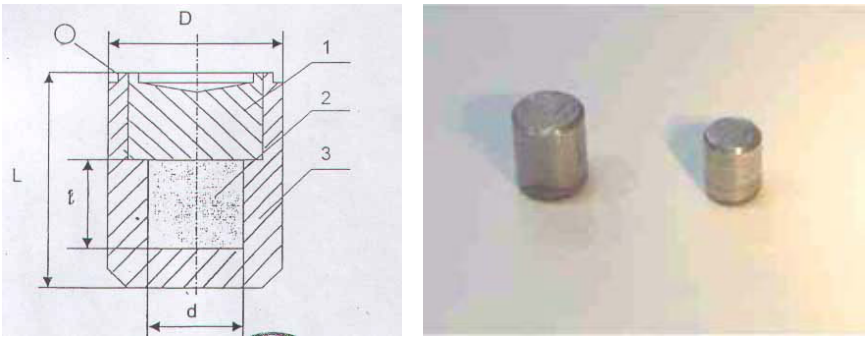

FIG. 5. Industrial ^{192}Ir source: 1 - plug, 2 -active core, 3 – capsule

TABLE IV. WELDING REGIMES FOR SOURCES TYPE GI192M54 AND GI192M55.

Type of source	Current strength, A	Rotate speed, revolution	Overhang of capsule from tong, mm	Arc distance, mm	Argon consumption, l/min
GI192M54	13-14	3.0-3.3	0.5	0.8-1.2	4-10
GI192M55	13-14	3.0-3.3	0.5	0.8-1.2	4-10

Sealing was performed by means of argon –arc circular edge weld. There were used following welding accessories and materials [11]:

- Power supply TRITON 160DC
- Type A argon (GOST 10137-79)
- Tungsten electrode type WT20 with diameter 2-3 mm and tool angle 15-25°.
- Cooper cooler tong.

The quality of welds depends in many respects of such preparing operations as

- Boiling in 10% solution of trilon-B, t=10 min.
- Boiling in distilled water, t=10 min.
- Washing in alcohol.
- Temperature drying 110-120° C, t=10 min.

The welding experiment consists of the following steps:

- Preparing the dummy sources accessories for welding;
- Assembling and sealing,
- Visual inspection
- Testing the air tightness by vacuum-liquid method;
- Microsection investigations.

The visual inspection of the welded samples shows the absence of any welding defects such as craters, pores, cracks and others.

Dimension inspection was done:

- the diameters of capsules in welding points are correspond with the technical specification for sources (4 mm for GI192M54 and 5 mm for GI192M55) (Table V);
- the heights of capsules are corresponding with the technical specification for sources (4 mm for GI192M54 and 5 mm for GI192M55) (Table V).

The air tightness tests show the absence of welding defects, too. Microsections of the tested sources are shown on Fig 6 and Fig 7.

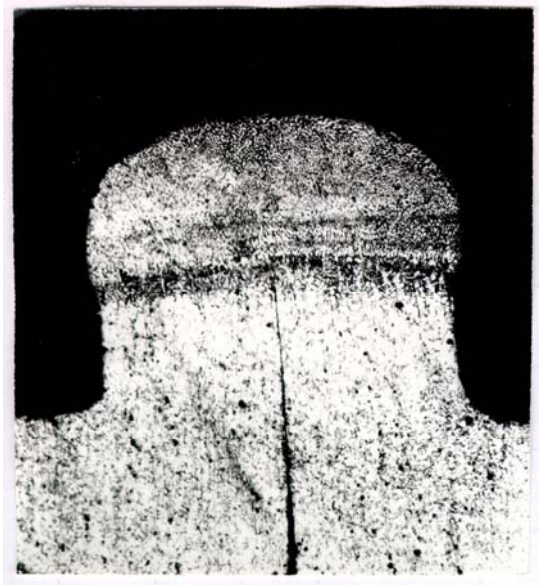

FIG. 6. Microsection of welds for GI192M54 capsule.

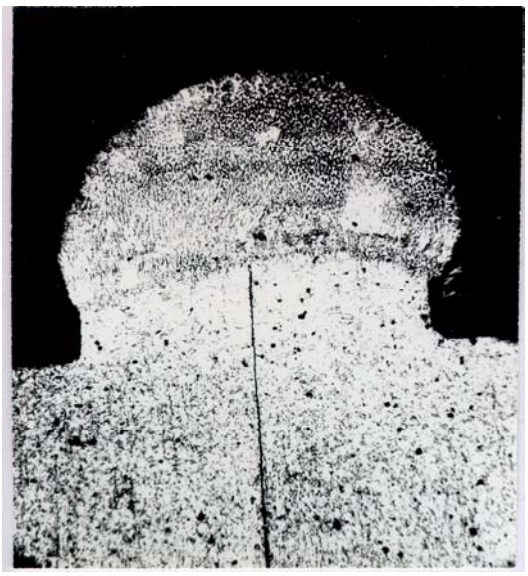

FIG. 7. Microsection of welds for GI192M55 capsule.

The microsection inspection of welded samples shows the absence of welding defects. The depth of welds is not less than 100 % of welded edges thickness. The structure of welds is austenitic. The growth of welded core is not essential.

TABLE V. SPECIFICATIONS FOR INDUSTRIAL SOURCES.

Source type	Dimensions, mm				Gamma dose rate at a distance of 1m, A/kg	Equivalent activity (estimated value) Bq(Ci)
	Source		Active Core			
	Diameter D	Length L	Diameter d	Length L (max)		
GI192M51	$4_{-0.022}$	5±0,15	0,5	0,5	(1,7-3,3) 10^{-8}	(1,8-3,7)x10^{10} (0,5-1)
GI192M52			1,0	1,0	(0,6-2,4) 10^{-7}	(0,7-2,6)x10^{11} (2-7)
GI192M53			1,5	1,5	(2,7-7,3) 10^{-7}	(3,0-8,0)x10^{11} (8-21)
GI192M54			2,0	2.0	(0,8-1,6) 10^{-6}	(0,9-1,7)x10^{12} (25-45)
GI192M55	$5_{-0.022}$	6±0,15	2,5	2,5	(1,8-2,6) 10^{-6}	(2,0-2,8)x10^{12} (55-75)
GI192M56			3,0	3,0	(2,8-4,2) 10^{-6}	(3,1-4,6)x10^{12} (85-125)
GI192M56-1			2,0		(0,8-1,7) 10^{-6}	(0,9-1,7)x10^{12} (25-45)
GI192M56-2			1,5		(2,7-7,3) 10^{-7}	(3,0-8,0)x10^{11} (8-21)
GI192M57	$6_{-0.022}$	7±0,15	3,5	3,5	(4,6-5,6) 10^{-6}	(5,0-6,1)x10^{12} (135-165)
GI192M58			4,0	4,0	(0,6-1,0) 10^{-5}	(0,7-1,1)x10^{13} (200-300)

TABLE VI. ASSESMENT OF RELATIVE UNCERTAINTY OF AIR KERMA RATE FOR IR-192 BRACHYTHERAPY SOURCES

Step	Type of physical quantity or procedure	Uncertainty (%)
1. Standards Laboratory.	Calibration of the secondary standard at SSDL	0.5
	Long term stability of the secondary standard	1.2
	Calibration of the user dosimeter at the standards lab	2.0
	Combined uncertainty in Step 1	2.39
2. Phantom Measurements	Long-term stability of user dosimeter	1.2
	Establishment of reference conditions	1.0
	Correction for influence quantities p_i and f_i	1.0
	Beam quality correction	1.0
	Combined uncertainty in Step 2	2.11
	Combined standard uncertainty	4.5

TABLE VII. ASSEMENT OF RELATIVE UNCERTAINTY OF AIR KERMA RATE FOR ^{192}Ir INDUSTRIAL SOURCES.

Step	Type of physical quantity or procedure	Uncertainty (%)
1. Standards Laboratory.	Calibration of the secondary standard at SSDL	0.5
	Long term stability of the secondary standard	1.2
	Calibration of the user dosimeter at the standards lab	2.0
	Combined uncertainty in Step 1	2.39
2. Phantom Measurements	Long-term stability of user dosimeter	1.2
	Establishment of reference conditions	3.0
	Correction for influence quantities p_i and f_i	3.5
	Beam quality correction	1.0
	Combined uncertainty in Step 2	4.87
	Combined standard uncertainty	7.26

There are determined preferable welding regimes for sources type GI192M54 and GI192M55 (Table III). As we can see from the table, for both type of capsules absolutely the same regimes. So we can apply this technology for manufacturing sources type GI192M51-M53 and GI192M56.

3.3. Quality control programme

The programme is extended to sealed wire ^{192}Ir sources of gamma-radiation with activity up to 10 Ci (370 GBq) and for industrial sources with activity ^{192}Ir up to 150 Ci (5.55×10^3 GBq). For manufacturing sources the supplies and materials have to pass inspection tests according to State Standard 24297-87 [14]. They should have certificates for their identification. Sources must be hermetically sealed. Leakage tests should be provided in accordance with immersion method 5.1.2 (ISO 9978/1992 [10]). The measured activity of liquid where sources have been treated must not exceed 185 Bq. Taking into account of short life of the source a periodical control is not provided for.

Estimation of source (or simulated sealed source) leak tight and strength must be carried out according to the results of examination before and after carrying out tests. Not less than two sources of the type specified are to be subjected to each test. The specific choice of sources is determined with the programme of tests. The programme of tests is developed according to ISO 2919:1999E [10] and State Standard 25926-90 [15] and is coordinated with the customers. When changing the design and the technology for manufacturing the source of the type specified which influence on its safe application as to purpose, new sources must be tested.

3.3.1. Tests execution

a. External examination

External examination should be fulfilled by visual inspection giving particular attention to quality of welding joints: the plug and the capsule, the wire and the capsule, the wire and the tail. Correspondence of dimensions of the source assembled with the requirements to specifications is checked with the certified gauges ensuring measurement accuracy specified by drawings.

b. Temperature tests (heating and cooling tests).

- Testing should be performed in heating and cooling devices.
- Heating is done in the devices providing with the temperature of the source up to +600 ^{0}C (+400 ^{0}C for industrial) and is lasted at this temperature not less than 1 hour. Then the source is subjected to testing by thermal shock. After heating the source should be transferred into running and non-running water with the ambient temperature not more than 20 ^{0}C for the time not more than 15 seconds.
- Consumption of running water per one minute must exceed the volume of the source by 10 and more times. The volume of non-running water must exceed the volume of the source by less than 20 times.
- Cooling of the source is fulfilled using solid carbon dioxide. The source is lasted at the temperature of –40^{0}C not less than 20 minutes.

c. External pressure tests

The source is placed into the chamber and it is subjected to pressure with air with the value of 2 MPa 5 minutes. Just the same testing is necessary in order these articles to be subjected to pressure of 25 kPa.

d. Impact tests

- For carrying out tests it is necessary: hammer with the mass of 50 g (5 kg for industrial) and the diameter of section of flat striking surface of 25 mm, with it outer edge rounded to radius of 3 mm. The center of gravity of the hammer lays on the axis of the circle which defines the striking surface; this axis itself passing through the point of attachment.
- The height of hammer falling down should be adjusted not less 1 m between the supposed surface of source collision arranged on steel anvil with the mass not less 500 g (5 kg for industrial) and impact surface of the hammer being in the starting position before free falling down.
- The source must be placed on the anvil horizontally in the rigidly fastened equipment – special jack.

e. Endurance test (for brachytherapy sources)

- Welding joints between the source and the wire, as well as between the wire and the shank must resist the load on rupture not less 15H.
- For carrying out tests it is necessary the source to be fastened vertically into a draw-in attachment, the calibrated weight with the mass of 1.5 kg is fastened with the same attachment to the shank.
- Rigidity rate of the test specified must coordinate with the customer.

f. Life time test (for brachytherapy sources)

- For carrying out this testing it is necessary to use the apparatus of contact radiation therapy or the device imitating it.
- Under the guaranteed number of sending of the source (1 from 100) must be subjected to not less than 5000 sending.

g. Puncture tests (for industrial sources)

- 300 g steel hammer, the upper part of which is equipped with means of attachment, and the lower part of which bears a rigidly fixed pin. The characteristics of pin are follows: hardness 50-60 Rockwell C; external height 6 mm; diameter (3±1) mm; string surface is hemispherical. The centreline of the pin is in alignment with the centre of gravity and with the point of attachment of the hammer.
- More than 3 kg hardened steel anvil with a cradle for source.
- The hammer is dropped from the height of 1 m onto the top of sealed source by means the smooth vertical tube. The pin is directed as close as possible to the welding joint.

The results of tests are considered negative if one of two sources after tests is found not sealed. The results of tests are considered positive if the source after tests preserves its tightness. The brachytherapy sources shall correspond to the ISO code C53211, and the industrial source shall correspond to the ISO code C43515 (ISO-2919:1999(E) [9]).

3.3.2. Measuring method

Method allows to calibrate sealed radioactive source with measurement error not exceed ± 5%. We are using for this purpose dosimeter “UNIDOS PTW Freiburg” or another device with same specification. The dosimeter should be calibrated by means of ^{60}Co sealed sources. The dose rate range is 1 μGy/min – 100 Gy/min. The ionization chamber Farmer 30001 PTW Freiburg is recommended as the main detector.

The following tools are recommended as auxiliary equipment.

- Cylindrical phantom type 9193 D=200 mm, H=120 mm with five holes (d=20mm) for measuring chambers and catheters.
- Thermometer, barometer.

In case of industrial sources special inserts are used. Taking into account relatively high activity of such kind of sources, up to 200 Ci (7.4×10^3 GBq), the high source surface temperatures for phantom material are applied. Insert is made from stainless steel. This insert also helps to place the source on axis with ionization chamber measuring point.

The ionization chamber with adapter is put onto central hole. Catheter with source is put into the one of the five side holes, other holes are closed by PMMA cylinders. Dose is measured placing the source in each hole. After that we measure background dose five times in same phantom.

Than we calculate arithmetic mean of dose measuring results:

$$\left(M_{uncor} \cdot p_t \cdot p_p\right)_R = \frac{1}{5}\sum\left(\left(M_{uncor} \cdot p_t \cdot p_p\right)_i - \left(M_{uncor} \cdot p_t \cdot p_p\right)_b\right) \quad (1),$$

where

- $\left(M_{uncor} \cdot p_t \cdot p_p\right)_i$ is dose measured with source;
- $\left(M_{uncor} \cdot p_t \cdot p_p\right)_b$ is background dose.

The air Kerma rate calculated as follow:

$$K_{ref} = \frac{\left(\left(M_{uncor} \cdot p_t \cdot p_p\right)_R\right) \cdot p_{hum} \cdot p_{ion} \cdot p_{pol} \cdot p_r \cdot p_u \cdot f_{np} \cdot f_{sc} \cdot f_{geo} \cdot d^2}{t} \cdot \quad (2),$$

where

- M_{uncor} is uncorrected dosimeter indication, microGy;
- p_t is correction factor for temperature;
- p_p is correction factor for pressure;
- p_{hum} is correction factor for humidity;
- p_{ion} is correction factor for ion recombination effect;

$$p_{ion} = \frac{\left(V_1 / V_2\right)^2 - 1}{\left(V_1 / V_2\right)^2 - M_1 / M_2} \quad (3),$$

where

- V_1 is usual ionization chamber voltage:
- V_2 is reduced ionization chamber voltage , V (it is recommended $V_1 \geq 3 \cdot V_2$);
- M_1 is reading obtained with the usual ionization chamber voltage;
- M_2 is reading obtained with reduced ionization chamber voltage.
- p_{pol} is correction factor for polarization effect;

$$p_{pol} = \frac{\left(\left|M_1\right| + \left|M_2\right|\right)}{2 \cdot \left|M_1\right|} \quad (4),$$

where

- M_1 is reading obtained with usual polarity;
- M_2 is reading obtained with opposite polarity.
- p_r is correction factor for filling the phantom material by ionization chamber;
- f_{wp} is correction factor for electron density difference between water and phantom material;
- f_{sc} is correction factor for difference between the value of scattered radiation in complete diffusion phantom and phantom PTW 9193;
- f_{geo} is correction factor for absorption and radiation scattering in water;

- p_w is correction factor for absorption and radiation scattering in chamber's walls and cap during calibration;
- d is distance from the center of chamber and to catheter axis, 0,08 m;
- t is measuring time, h (t ≥ 1 min).

It can be calculated:

$$C = p_{hum}\, p_r p_w f_{wp}\, f_{sc}\, f_{geo}\, d^2 = 0.0075 \qquad \text{(for } ^{192}\text{Ir) [6, 7, 8].}$$

So

$$K_{ref} = \frac{\left(\left(M_{uncor} \cdot p_t \cdot p_p\right)_R\right) \cdot p_{ion} \cdot p_{pol} \cdot C}{t}. \tag{5}$$

Activity of source we calculate as follow:

$$A = \frac{K_{ref}}{\Gamma_\delta} \tag{6}$$

where

- Γ_δ is constant for ^{192}Ir = 0,1137 μGy м^2/h·MBq.
- The relative uncertainty is shown on Tables VI and VII [13].

4. CONCLUSIONS

The sealing method for brachytherapy afterloading sources were developed by means laser-welding system. Different welding regimes were investigated. The mechanical and optical parts of welding system were adjusted to obtain a better quality of welding joints. The sealing method for industrial sources was developed by means argon arc welding. Pilot batch of dummy sources were produced and investigated. The QC programme was developed for brachytherapy and industrial sources. The QC programme consists of external examination, temperature tests, mechanical tests and measuring method. The Special Form Radioactive Material certificates were obtained for each kind of sources.

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PREPARATION OF ^{125}I SEED SOURCES FOR IMPLANTATION THERAPY OF PROSTATE CANCER

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Abstract

The scope of the research was to optimize the adsorption of ^{125}I on core seeds using radiochemical techniques. The encapsulation, the Ag bar uniformity adsorption and self-shielding of ^{125}I seed source were investigated. The chemical processing and TIG (tungsten inert gas) welding technology of ^{125}I seed source bar were studied. Silver bar chlorination, iodination and self absorption of ^{125}I were quantified; for example, reagent selection of chlorination and iodination, reagent quantity, reaction time, processing bar, reaction medium, reaction rate and temperature were measured. The quality control of ^{125}I seed source was performed. The adsorption efficiency of ^{125}I in a large batch production was found very high. The homogeneity and reproducibility of ^{125}I seed source between batch and intra-batch was considered optimal.

1. INTRODUCTION

The medical ^{125}I seed source consists of silver bar ^{125}I source (0.5x3.0mm) and titanium encapsulation shell (0.8x4.5mm). ^{125}I brachytherapy sources have a half-life of 59.4 days; they decay by electron capture with the emission of Auger electrons generating 27.2 - 31.9 kV X rays and a gamma emission of 35.5 keV. The electrons are absorbed by the titanium wall of the capsule. These sources can be used either as primary treatment or for treatment of residual disease after excision of the tumor. Seeds in the apparent activity range of 0.1-1.0 mCi (3.7 – 37 MBq) can treat the superficial, intraabdomiral, and intratoracil tumors; tumors of the head, neck, lung, pancreas and prostate (early stages) are commonly treated. ^{125}Iseed sources have been used largely in the clinical therapy in Europe and America. It is already recognized their clinical benefit.

2. SCOPE OF PROJECT

The scope of the research was to optimize the adsorption of ^{125}I on core seeds using radiochemical techniques. The encapsulation, quality assurance method, the Ag bar uniformity adsorption and self-shielding of ^{125}I seed source were investigated. The following are steps of the investigation:

(1) The quality control of silver bar
(2) Silver bar chlorination
(3) Silver bar iodination
(4) Ag bar uniformity adsorption
(5) Self-shielding of ^{125}I seed source

3. RESULTS AND DISCUSSION

3.1. The quality control of silver bar

Quality of silver bar (size, weight, and appearance) affects directly chlorination and iodination, so it needs different control methods. The measurement error of silver bar has been in range of 3%.

3.1.1. Silver bar chlorination

Sodium hypochlorite, hydrogen dioxide solution and hydrochloric acid were different systems for oxidized and chloridized silver bar. Sodium hypochlorit and hydrochloric acid system produced a lot of Kelly gas (Cl_2) when it chloridized silver bar. But hydrogen dioxide solution and hydrochloric

acid system produced a little chlorine gas when hydrochloric acid was used. When hydrogen dioxide solution and hydrochloric acid ratio, chlorinated time and vibration speed are controlled, chlorine gas is not produced, silver bar surface was uniformity and compact (Tables I,II, III).

TABLE I. HYDROGEN DIOXIDE SOLUTION AND HYDROCHLORIC ACID SYSTEM

Volume ratio	5	1	0.2
NaClO/HCl（6N）	Have Kelly gas, Uniformity, compact	Have Kelly gas, Uniformity, compact	Have not Kelly gas, Asymmetry, shagginess
H_2O_2/HCl, 6N	Have not Kelly gas, Uniformity, compact	Have not Kelly gas, Uniformity, compact	Some Kelly gas, asymmetry, shagginess

TABLE II. VIBRATION TIME EFFECTS ON CHLORINATED BAR

Vibration time（min）	10	30	60	90
Quantity of chlorination	Rather uniformity, Shagginess	Rather uniformity and compact	Uniformity, Compact	Uniformity, Compact

TABLE III. VIBRATION SPEED EFFECTS ON CHLORINATED BAR

Vibration speed, N/min	20	50	100
Quantity of chlorination	Uniformity, Compact	Uniformity, Compact	Rather uniformity and compact

3.1.2. Silver bar iodination

^{125}I adsorption rate depends on various parameters, such as adsorption time, adsorption volume, ^{125}I activity concentration, adsorption temperature, pH value, quantity of carrier. Figure 1 shows the adsorption rate versus these parameters.

3.2. ^{125}I - Ag bar uniformity adsorption.

^{125}I-Ag bar uniformity adsorption was investigated. The optimum of silver bar iodination was fixed changing the quantity of silver bar. When the reaction was finished, the adsorption rate of silver bar was measured. The results showed that the adsorption rate decreased when the quantity of silver bar increased, while the average error of silver bar increased continuously. The number of ^{125}I-Ag bar uniformity adsorption is presented in the Table IV.

TABLE IV. ^{125}I-AG BAR UNIFORMITY ADSORPTION

Number（N）	2	4	6	8	10
Adsorption rate（%）	91	90	88	75	68
Average error (σ)	3.21	4.13	5.12	5.64	6.36

The optimum of the silver bar iodination were investigated changing the quantity of the carrier. When reaction was finished, the adsorption rate of the silver bar was measured. The results showed that the quantity of carrier affects the ^{125}I-Ag bar uniformity adsorption; when the quantity of the carrier increases, the average error of silver bar becomes smaller. The effect of the quantity of carrier on ^{125}I-Ag bar uniformity adsorption is presented in the Table V.

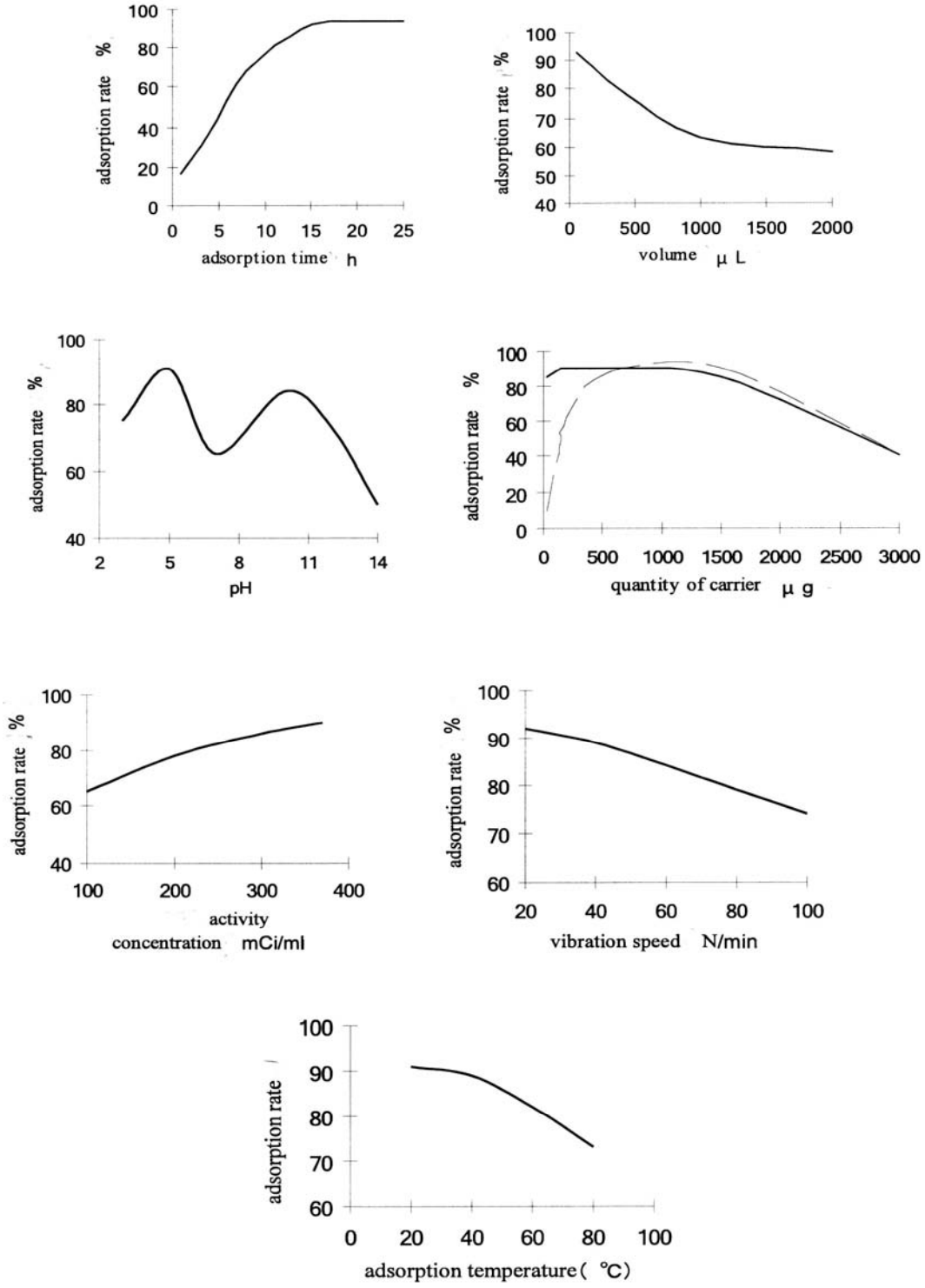

FIG. 1. The adsorption rate of ^{125}I versus adsorption time, volume, pH, quantity of carrier, activity concentration, vibration speed and adsorption temperature.

TABLE V. QUANTITY OF CARRIER AFFECTS ON ^{125}I-AG BAR UNIFORMITY ADSORPTION

Carrier content, μg/ml	6	15	24	30	60	150
No.1 activity,MBq	20.3	20.0	22.2	18.3	20.0	20.3
No.2 activity, MBq	31.3	22.9	28.4	23.6	21.7	20.7
No.3 activity, MBq	14.4	16.2	25.1	22.2	21.0	19.2
Average error (σ)	8.58	3.36	3.10	2.75	0.85	0.78

3.3. Self-absorption of ^{125}I seed source

3.3.1. Self-shielding of ^{125}I-Ag bar

Put iodinated silver bar into mixed solution of sodium cyanide and sodium hydroxide. ^{125}I was dissolved in the medium from iodinated silver bar and measured activity, activity of solution and iodinated silver bar surface activity differentiated value ratio activity of solution was defined self absorption of iodinated silver bar. The results of self-absorption of ^{125}I-Ag bar showed the effect quantity of carrier were little. Self-absorption of ^{125}I-Ag bar was close to 40 percent (Table VI).

TABLE VI. SELF-SHIELDING OF ^{125}I-AG BAR

Carrier content（mg/ml）	3	6	15	24	30	60	150
Apparent activity of ^{125}I-Ag (MBq)	19.8	20.3	20.0	22.0	18.3	20.0	20.3
Calculation activity of ^{125}I solution (MBq)	31.6	34.2	32.5	36.4	29.6	32.5	33.3
Self-absorption (%)	37.3	40.7	38.6	39.5	37.7	38.4	39.1
Average self-absorption (%)	38.8						

3.3.2 Titanium tube shield

Apparent activity of ^{125}I seed source and ^{125}I-Ag bar were measured, then titanium tube shield rate calculated. Average shield rate was close to 9 percent. Total self-shielding of ^{125}I seed source were close to 40-50 percent (Table VII).TITANIUM TUBE SELF-SHIELDING

Apparent activity of ^{125}I-Ag (MBq)	36.2	42.3	44.2
Apparent activity of ^{125}I seed source (MBq)	33.1	38.8	40.3
Shield rate（%）	8.6	8.3	8.8
Average self-shielding rate （%）	8.6		

Figure 2 shows the self-develop phenomena of ^{125}I seed source.

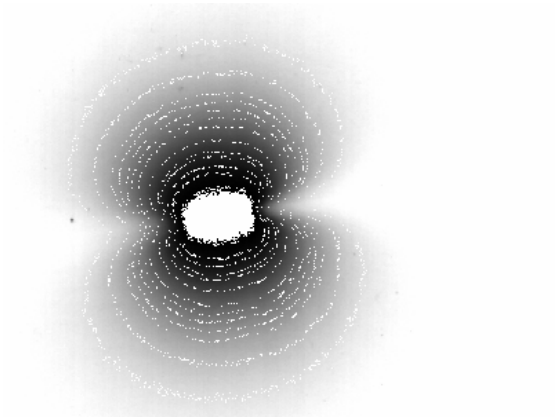

FIG. 2. Self-develop of ^{125}I seed source

4. CONCLUSIONS

The results of ^{125}I adsorption research of chlorinated bar have shown that the optimal regime of silver bar iodination is the adsorption time over 15 h and pH value controlled at 5. The ^{125}I adsorption rate is high when quantity of carrier and ^{125}I activity concentration increases. It is very important to control the adsorption temperature and the adsorption volume. The carrier quantity on ^{125}I-Ag bar uniformity adsorption was increased and the average error of silver bar became very small. Total slf-shielding of ^{125}I seed source were close to 40-50 percent.

The research work proposed for the near future is the following:

1. Establish quality control seed source (radioactive activity of every seed source, isodose curve, superficial contaminated and leakage, etc.)
2. Animal test
3. Preliminary clinical trial

NEW METHODS IN MINIATURE TECHNIQUES FOR PREPARATION OF SEALED SOURCES FOR MEDICAL APPLICATION

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Abstract

The aim of the research project was the study of new methods for miniature sealed sources for medical applications based on reactor produced radionuclides and investigation of sealing technology. Reactor produced radionuclides investigated were: ^{32}P, ^{90}Y, ^{125}I, ^{153}Sm, ^{166}Ho, ^{192}Ir. The most widely used radionuclide for brachytherapy treatment purposes is ^{192}Ir. The laser welding equipment designed and produced in Hungary is sufficient to weld stainless steel flexible wire ropes on the horizontal position and inactive circumstances but the beam current is not enough for titanium capsule to be welded on the vertical position without damaging or burning the capsule. IZOTOP's manufacturing operates in accordance with the ISO-9001 quality assurance system. All products are subject to stringent safety measures and controls. Environmental protection against radioactive contamination is ensured by a multi-level safety system.

1. INTRODUCTION

Radiation sealed source production was started at the Institute of Isotopes more than 30 years ago. The production technologies have been developed and used routinely of ^{60}Co gamma sources for industrial and medical, ^{192}Ir gamma sources for NDT purposes. Radiation sealed source production in Hungary was based on the Research Reactor in Budapest (^{60}Co low activity), the cobalt target irradiation for industrial irradiator is used NPP in Paks and activation of iridium pellets is performed in Petten's reactor.

Radiation sealed sources are manufactured using a special hot-cell system for ^{192}Ir and for ^{60}Co. The high capacity hot-cell system has been also developed and built up for production of high activity ^{60}Co sealed sources. There is a variety of specialized computer controlled remote after loading system like a type GM 12i. This system is using a high activity point source of ^{192}Ir. Iridium-192 in the form of wire is used for treatment of tumour in breast, tong etc. Institute of Isotopes Co. Ltd. has been developing the ^{192}Ir sources for this purpose.

Brachytherapy is a method of medical radiotherapy where a sealed radioactive source is transferred close to the target tissue using an applicator. The after loading equipment is being increasingly utilized in the cancer treatments.

2. MATERIALS AND METHODS:

2.1. Development of the ^{192}Ir medical sealed sources for HDR afterloading system

2.1.1. Determination of the irradiation for Iridium target

- Physical form of target: metal disc: (∅0,5x0,5 mm) or wire: (∅0,6 mm)
- Specific activity of the irradiated target: min. 18,5-20,4 TBq/g (450-550 Ci/g)

Technical information of the source:

- Active dimension: ∅0,6x3,5 mm
- Inactive dimension: ∅1,1x5,1 mm (+ 2100 mm flexible wire rope)
- Activity: max 555 GBq
- Capsule material: stainless steel (KO 36 or DIN 1.4541)
- Drawing: 48H-01
- Type of sources: IrS-48H, IrS-48H-2

2.2. Investigation of sealing technology

The advantage of Laser welding technology is that during the welding procedure the thermal effect for mechanical characteristics will be the same as before. There are three beams for different optical functions: welding beam, aiming beam and observer beam. Parameters of the laser welding system were:

- Diameter of laser beam: max 120 μm
- Impulse: 3-5 μsec
- Output: 10 mJ
- Wave length: 1064 nm

Main units of the optical system were:

- TV camera
- Objective
- Mirror (dichroical)
- Deflecting mirrors
- Tube lense
- Focusing objective
- Working laser (Nd:YAG)
- He-Ne laser for aiming beam
- Lighting
- Target to be welded

2.2.1. Determination of the mechanical system

- Stable basement for supporting the parts of system (should be insulated against vibration) Dimensions: 500x200x10 mm steel plate
- Rotating unit, adjustable in 3D (400 steps/rot)
- The target to be welded can be positioned and rotated around the longitudinal axis with a special mechanical system.

2.2.2. Determination of the control system

The control system contains: IBM AT 486-DX4, 120 MHz PC, 16 MB RAM, 840 MB HD, 1,44 MB FDD, SVGA 17" Monitor, SVGA graphic card, FRG 2M frame grabber card, STEP-52 3 channel step-motor control cards, mouse, joystick. PC controlled system which provides the following controls:

- Control of required motion
- Control of target rotation
- Control of laser-impulse
- Display of the TV camera image
- Control of adjusting
- Control of welding parameters
- Control of automatic welding process

2.3. Main procedure of the miniature source production

2.3.1. Preparation of the inactive assembly

- Producing of the source capsules, cap and conical end with turning machine using stainless steel rod.

- Producing of the stainless steel wire ropes using micro plasma welding equipment with the following parameters: dia of nozzle 1 mm, Pilot current: 5 Amps, Welding current: 3,5 Amps, Plasma gas: 0,2-0,4 l/min, Inert gas 10 l/min, Welding time: 0,4 sec.

2.3.2. Assembling of the sources

The active row material to be transported in a B(U) type container should be loaded into the assembled inactive parts using an appropriate vessel and trays.

2.3.3. Welding of the sources

- The welding should be carried out with a special holder unit and a micro plasma welding equipment type Plasmafix 50E with the following parameters:
    - diameter of nozzle 1,2 mm,
    - Pilot current: 2 Amps,
    - Welding current: 1,5 Amps,
    - Plasma gas: 0,3 l/min, Inert gas 8 l/min,
    - Welding time: 0,8 sec.
- Visual checking of the welding quality.

2.3.4. Testing of the surface contamination and leakage:

- Using ultrasonic test with solutions and measuring the activity of the solutions.
- The source is sealed and free from contamination if the activity of the solution is less than 185 Bq.

2.3.5. Activity measurement of the sources:

- Check the measuring system using the reference source. In the hot cell during activity measurement should be only one, the actual source to be measured.
- The source activity measurement should be carried out with an ionisation chamber detector.

2.3.6. Certification of sources

About each sealed source should be written a “Source Certificate”. This certificate contains the most important significant data of the sealed source concerning the International Requirements.

2.4. Quality assurance and safety

Quality Assurance (QA) and Quality control (QC) are two terms which are often confused. QA is the total process whereby a manufacturer ensures that a product meets the quality required for its intended use. It covers the design and development stages through to manufacturing, quality control and delivery process to final storage and use by the customers.

Quality cannot be controlled into a product after it is made. It is essential to design in the right qualities at the development phase so that the manufacture, quality control and use by the customers are easily and uncomplicated.

The QA process at this time will be directed at both product and regulatory needs and will include the establishment of documented support for product claims, testing of routinely produced batches stored under normal and stress conditions for setting of specifications, validation of manufacturing and quality control processes as well as extensive clinical trial and safety support investigations.

All of IZOTOP's manufacturing and other activities operate in accordance with the ISO-9001 quality assurance system. The first certificate was granted in April 1998. Audits performed by SGS International are regularly updated. In addition, the production of in-vivo products also follows the principles of GMP as licensed by the National Institute of Pharmacy.

All of IZOTOP's products are subject to stringent safety measures and controls. Environmental protection against radioactive contamination is ensured by a multi-level safety system.

2.4.1. Quality control of medical radiation sources

Quality assurance

Radiation sources are manufactured in accordance with a strict quality assurance programme, detailed of which can be obtained on request.

Testing for leakage and contamination

Stringent tests for leakage are an essential feature of radioactive source production. The methods adopted depend on the design and intended application of the source, and also on statutory requirements. Where necessary, tests can be specially modified to meet particular requirements. Checks made routinely during production

The standard methods used for testing radiation sources are listed below:

- Wipe test A
- Bubble test D
- Immersion test L
- Helium leak test H

Test report:

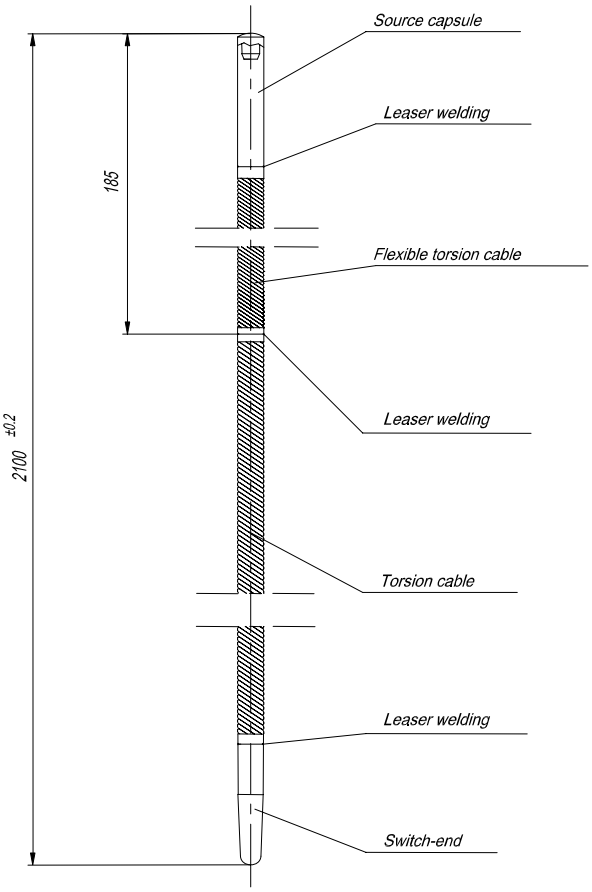
- A test report is supplied with each source or batch of sources.
- Product code
- Product description
- Capsule type
- ISO classification
- Special form certificate
- Serial number of source
- Measurement check
- Contamination check
- Leak test

3. CONCLUSIONS

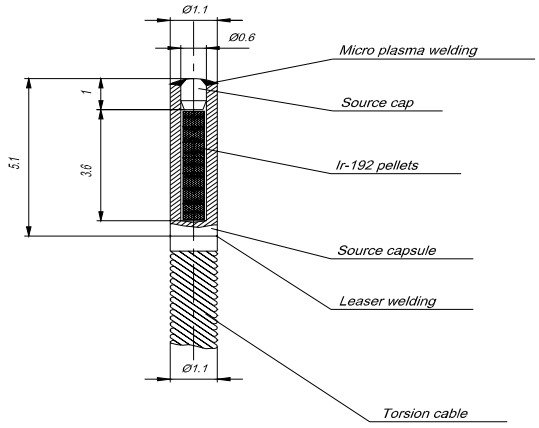
The laser welding equipment designed and produced in Hungary is sufficient to weld stainless steel flexible wire ropes on the horizontal position and inactive circumstances but the output power (beam current) is not enough for titanium capsule to be welded on the vertical position without damaging or burning the capsule.

Investigations on the upgraded laser welding equipment and technology to seal titanium encapsulated sources for medical application are going on as follows:

- Determination of the new parameter of welding technology
- Positioning and visual control system to be installed into the hot cell.
- Investigations on the Quality control procedures and equipment.



IrS-48H



IrS-48H

FIG. 1. ^{192}Ir sources

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DEVELOPMENT OF MINIATURE ^{125}I SOURCES FOR THE TREATMENT OF EYE AND PROSTATE CANCERS

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Abstract

The aim of the present work was to develop a technology for the fabrication of ^{125}I brachytherapy sources for their potential application in the treatment of eye and prostrate cancers. Several methods of incorporating the ^{125}I activity in solid core were investigated and the best among them, in terms of low leachability, ease of preparation & encapsulation in titanium capsules and reproducibility was chosen. Two kinds of cores, namely rod/cylinder shaped wires and spherical balls, were envisaged for introduction into the titanium capsule. Two procedures for depositing the ^{125}I activity on rod shaped core were explored; namely anodic deposition of ^{125}I on silver wire and physicochemical adsorption of ^{125}I on palladium chloride coated silver wire. Similarly, two methods for depositing ^{125}I on spherical source cores were studied. In the first case, alumina microspheres were used as the core on which ^{125}I was impregnated as iodate (IO^-_3) using a solid-solution interface technique. In the second, ^{125}I was adsorbed on palladium coated silver beads of 0.5 mm. Various physical and chemical parameters were optimized to obtain quantitative uptake of ^{125}I and firm adsorption that would result in negligible iodine leachability (≤ 0.01%). The source cores prepared by various methods were encapsulated in indigenously fabricated titanium capsules [4.5mm (l) 0.8mm (OD) and 0.05mm (thickness)]. These capsules were in accordance with the ISO specifications for use as low energy brachytherapy sources. The welding of the capsules was optimized by changing welding parameters such as energy, frequency and pulse duration to obtain neat and leak-proof welded capsules. Integrity of the welding was ascertained by metallographic examination and leak testing. Quality assurance tests such as leak testing and dosimetry that are essential before clinical use were conducted. All the above methods were compared and the one which was easy to prepare in large scale was adopted for preparation of the sources for clinical trials.

1. INTRODUCTION

Interstitial brachytherapy using ^{125}I sources for the treatment of eye and prostate cancer is now an established radiation therapy modality. The attractive efficacy of treatment of certain kinds of ocular cancers such as retinoblastoma and choroidal melanoma by this mode has resulted in increased awareness and increased demand for these sources. Retinoblastoma is a relatively uncommon tumor of retina, occurring in babies and children younger than 15.

Choroidal melanoma is the most common primary intraocular (occurring inside the eye) tumor in adults. In both cases, the cancer may metastasize and eventually spread to other parts of the body. If ignored, intraocular tumor can cause protrusion of the eye ball and spread to other organs like brain etc. Although the primary modality of treatment has been chemotherapy and external beam radiotherapy, it is often seen that it is necessary to enucleate the eye which leaves the patient sightless (on one side if one eye is involved or total if both eyes are involved) for the rest of the life. It has been reported that complete remission may be obtained by treating with low energy radiation sources placed in contact with the tumour. Thus, plaque brachytherapy of ocular tumours has emerged as an effective method for control of localised tumours with eye preservation. On similar lines, low energy radiation sources, are also permanently implanted for clinical management of certain types of prostate cancers [1].

^{125}I is among the most commonly used radionuclides for treatment of both ocular and prostate cancers. Additionally, ^{106}Ru has been reported for treatment of ocular cancers while ^{103}Pd has been used in the case of prostate cancers. The amount of activity in each seed like source, the number of sources and the distribution of the sources for treatment will depend on the size of the tumour and the dose distribution desired for best efficacy, as decided by the radiation physicist/oncologist. In the case of ^{125}I, often 10-20 sources of 74-111 MBq (2-3 mCi) are used in ocular tumours while 40-100 sources of ~ 37 MBq (1 mCi) are used in prostate cancers.

Both ^{125}I (T1/2 ~60d, 27-32 keV Te X rays and 35 keV γ ray) and ^{103}Pd ($T_{1/2}$ = 17 d, 20-23 keV Rh X rays) have similar radiation characteristics. However, the long half life of ~60 days and feasibility of production in nuclear reactors makes ^{125}I a better option.

Development of radiation sources for ophthalmic plaque therapy was taken up by the Radiopharmaceuticals Division, BARC, at the request of Sankara Nethralaya, Chennai, India, one of the leading eye hospitals of India. The goal was to make ^{125}I- brachytherapy sources for the treatment of ocular and prostate cancers and supply to the oncology centers in India at affordable cost. At that juncture, this coordinated research which aimed at developing the technology for the production of ^{125}I and ^{125}I-brachytherapy sources was very apt for participation by our centre.

Iodine belongs to the halogen group of elements which is highly reactive. Although several stable compounds of iodides are reported in the literature, most of them have a definite solubility in water/saline. The main challenge, therefore, was to develop a non-leachable source core containing ^{125}I incorporated in a solid substrate at very high specific activity. Production of the source core in a highly reproducible manner within acceptable dimensional tolerances was yet another challenge. An innovative strategy had to be devised to develop the necessary technology for fabrication of ^{125}I source core, capsules and encapsulation technique. Some of the techniques and matrices used for making ^{125}I source core are electrochemical deposition, adsorption on organic materials, coating on to tungsten surface, adsorption on palladium wire, coating on silver and ceramic beads, coating on ion exchange resins, etc. Cieszykowske et al. have reported an electrochemical method of depositing ^{125}I on silver matrix [2].

The Model 3500 I-Plant ^{125}I brachytherapy source produced by Implant Sciences Corporation is created through a novel technique which utilizes ion implantation of ^{124}Xe into a ceramic matrix followed by neutron activation of the substrate [3]. Zhang et al. have studied the preparation of ^{125}I seeds using silver as carrier body and employing ion-exchange technique [4]. Han et al. studied the absorption of ^{131}I on a ceramic matrix which can also be used for the preparation of ^{125}I seeds [5].

The Model LS-1 BrachySeedTM , manufactured by M/s DRAXIMAGE Inc., consists of two ceramic beads impregnated with ^{125}I [6]. The Best® Model 2301, produced by M/s Best Medical International, ^{125}I source core comprises of a tungsten X ray marker coated with an organic carbon layer impregnated with ^{125}I [7]. The select seed ^{125}I prostate, marketed by M/s Nucletron Corporation, is created through a technique in which ^{125}I is adsorbed on to silver matrix as silver halide layer [8]. The ^{125}I activity can also be absorbed on an ion exchange resin bead [9,10,11] to make source core. But, the details of most of these procedures are covered by patent rights (IPR) and not disclosed. However, the dosimetric evaluation of these commercial sources are periodically investigated and reported in the literature [12-15].

At our laboratories development of ^{125}I-source seeds was undertaken addressing the following main issues:

- Immobilization of ^{125}I in a suitable solid matrix.
- Non-leachable characteristic of the matrix.
- Fabrication of Ti-capsules of suitable dimension.
- Hermetic sealing of the capsules by Laser welding.

The capsule matrix should have chemical compatibility with the source core which will be encapsulated in it, good strength, low weight and outstanding corrosion resistance properties. As titanium metal satisfies most of these characteristics, it was chosen as the capsule material. The outer dimension of the titanium encapsulated source for ocular and prostate radiotherapy, as specified by ISO, is 4.5mm (l) x 0.8mm (OD) with wall thickness of 0.05 mm (Fig. 1).

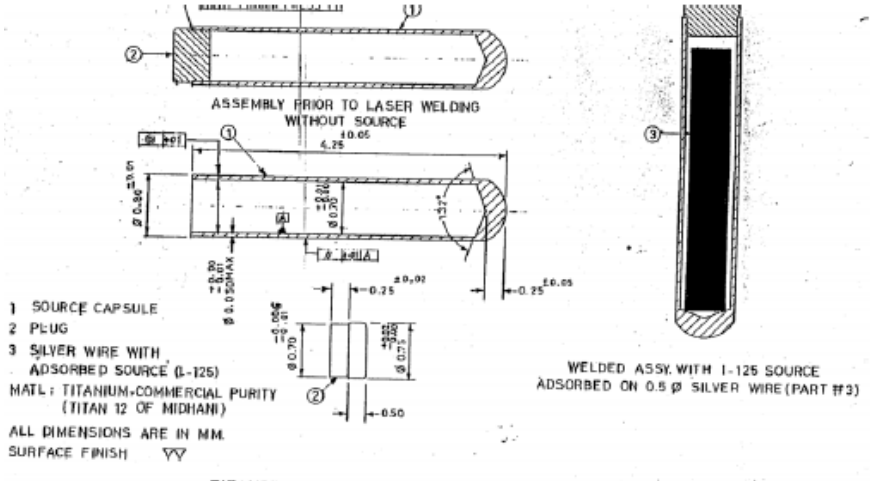


FIG. 1. Titanium capsules for ^{125}I brachytherapy sources

This work gives an account of the development of the sources, development of the capsules, laser welding the sources to meet the stringent safety parameters, qualification of the sources for human application and deployment for clinical trials.

2. MATERIALS AND EQUIPMENT

Silver wire and beads of 99.9 % guaranteed purity and platinum wire of 1mm (dia.) were purchased from reputed local commercial suppliers. Titanium rods used for the fabrication of capsules was purchased from MIDHANI (A public sector unit of India), Hyderabad, India. Titanium capsules were fabricated by Hindustan Machine Tools (HMT) in their Precision Machinery Division (A public sector unit of India). ^{125}I used for these studies was initially imported, but later produced in our reactors and processed in our Division. All other chemicals required for the work was obtained from reputed commercial suppliers. Ion chamber calibrated for ^{125}I was purchased from M/s Sun Corporation, Florida USA. Laser welding machine was obtained from Quanta systems, Italy.

3. METHODS

Two types of source core were studied; namely:

- Rod/wire type sources
- Spherical type sources

3.1. Preparation of rod/wire type sources

The following methodologies were investigated for preparing ^{125}I-silver wire source core:

- Electrodeposition
- Physical & physico-chemical adsorption

3.1.1. Electrodeposition

Anodic electro deposition of radio iodine (^{125}I) was carried out in a quartz bath size [1.2 cm (dia.), 2.5 cm (ht)] with platinum cathode (1mm). The wires were arranged in the cell as shown in Fig.2. Thoroughly washed silver wire was placed at the center of the cell as anode. Radio iodine solution 111-130 MBq (3-3.5 mCi)/30 µl was added to 0.01M Na_2SO_4, adjusted to pH 9 using 1mM ammonia and the total volume made up to 650µl.

The deposited radioactivity was measured using a dosimeter. Various experimental parameters such as the current used, radioiodine concentration in the cell and time for deposition were optimized to obtain maximum activity on the silver wire.

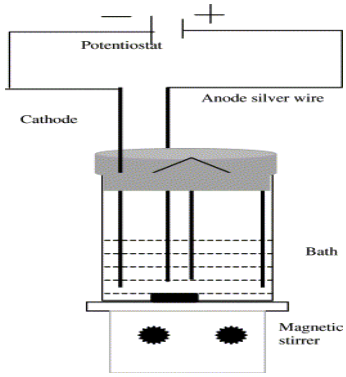


FIG. 2. Schematic diagram of the electro-deposition set-up.

3.1.2. Physical & physico-chemical adsorption

Pure silver wires (0.5mm diameter) cut into pieces of 3mm length were thoroughly washed with acetone followed by hot and cold water. They were further treated with 3 M HCl and washed free of acid with double distilled water and were completely dried under Infra red lamp to a constant weight and were used for adsorption experiments.

In order to effect chemical reaction leading to the immobilization of iodine on silver, coating of silver wires with a chemical which has affinity for iodine was considered. The pre washed silver wires were immersed in $PdCl_2$ solution at acidic pH. The solution was boiled for 5 min and cooled. The wires were thoroughly washed in double distilled water and dried to a constant weight.

In both cases (pure silver as well as Pd coated silver), the cleaned wires were individually immersed in a small conical shaped test tubes containing a mixture of ~148 MBq (~ 4 mCi) of ^{125}I and 5 μg of KI as carrier in a total volume of 50 μl. The adsorption experiments were carried out to optimize experimental parameters for quantitative adsorption of radio iodine on the silver wire. The effect of volume (50-500 μl) of radioiodine, presence carrier concentration (2μg-10μg), temperature (22°-70°C) during adsorption and time of contact (0.5-7 h) with radioactive solution on the percentage adsorption were studied. At the end of these experiments, the wires were washed thoroughly with double distilled and dried under infrared lamp. The approximate radioactivity on the wires was measured by a radiation monitor using dose vs. activity relationship at a fixed geometry.

A reported method based on absolute counting [17] was used to accurately measure the residual unadsorbed activity and washings from individual tubes. The percentage adsorption was deduced from these data.

3.2. Preparation of spherical type sources

Alumina microspheres and metallic silver spheres were used as spherical sources.

3.2.1. Alumina microspheres

Preparation of alumina microspheres

A mixture of pre-cooled (5°C) solution of hexamethylenetetraamine and urea (3M) with aluminum nitrate solution was dispersed as droplets into hot oil to bring about the conversion to the solid gel sphere form.

The spheres were separated from the oil and collected on a wire mesh, washed free of grease with carbon tetrachloride and then washed thoroughly with ammonia solution (2M) to remove the chemical impurities occluded inside the spheres. The spheres were dried in an air oven at 100°C and heat treated at 700°C for 5h in a furnace. The uniform sized spheres were selected by passing through a 600 μm mesh.

Method of coating the alumina microspheres with ^{125}I

^{125}I was immobilised on the microspheres by following a modified procedure reported earlier [19]. Five microspheres, previously conditioned with 1 mM sodium nitrate at pH 2–3, were immersed in ^{125}I (100 mCi/ml) followed by the addition of 0.1 ml of freshly prepared chloramine-T solution (1 mg/ml in 1 mM sodium nitrate) in a quartz test tube. The tube was closed tightly and the microspheres were shaken using auto shaker at ambient temperature (22°C). The spheres were washed twice in the 1 mM sodium nitrate to remove the unadsorbed activity and dried at room temperature. The percentage adsorption of iodine activity on the spheres was indirectly measured by counting the reminder of the adsorbing solution and the washings in a well-type scintillation counter set for 125I activity, after sufficient dilution. The adsorption experiments were carried out at different time intervals ranging from 1 to 5 h and at different radioactive concentrations to optimize the time and volume taken for maximum adsorption.

3.2.2. Metallic silver spheres (beads)

Method of coating of radioactivity on silver beads

Silver beads of ~ 0.5 mm (ϕ) were cleaned with 3M HCl followed by thorough washing with water. The coating of palladium on silver beads was carried out by treating the beads with $PdCl_2$ solution at ~100˚C for 15-20 minutes. The palladium coated beads were washed and dried to constant weight. The beads were individually taken in small glass reaction tubes and treated with ~10μL of ^{125}I solution containing ~ 9.25 MBq (250 μCi) of ^{125}I at pH 9-10. Carrier iodide as KI was added to the individual reaction tubes to enhance the adsorption kinetics. The total reaction volume was maintained as 15μL and the reaction temperature was optimized for quantitative adsorption. The adsorption was carried out at ~60-70ºC for 6-8 h. The adsorbed beads were washed with warm (~50ºC) distilled water and the percentage adsorption was estimated by measuring the radioactivity of beads using a well type NaI(Tl) scintillation counter set for ^{125}I. The activity in the bead after adsorption was measured by using a pre-calibrated ion chamber.

3.3. Adsorption studies

The adsorption of ^{125}I on the chemical treated wires/spheres was studied by varying the experimental parameters such as the volume of radioactive solution taken for adsorption, temperature and carrier concentration to optimize the adsorption and obtain quantitative yields.

3.4. Determination of uniformity of adsorbed ^{125}I activity

Uniformity of deposition of ^{125}I activity was examined by autoradiography using a specially designed gadget (Fig.3.) A circular disc (4.4cm dia., 1.4cm thick) made of brass with a central hole of 3mm diameter and 8 mm depth was taken and eight equidistant tunnels (45^o angle between each successive tunnel) of uniform aperture were drilled through the central hole. One source was placed at a time in the central hole and autoradiographed simultaneously by wrapping a strip of photographic film all along the side of brass disc. The film gets exposed from eight equidistant directions through the holes. The optical density distribution of the exposed film at different angles was measured by B/W transmission densitometer. Six randomly selected source cores were thus autoradiographed one after another.

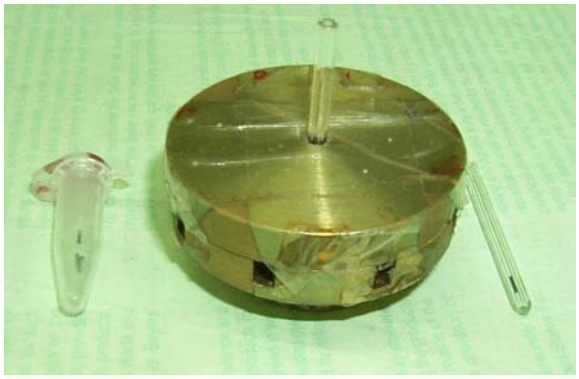


FIG. 3. Gadget for Auto-radiography

3.5. Studies on the leachability of the ^{125}I

The leachability test on the radioiodine deposited wires was carried out as per a method prescribed for finished sources by Atomic Energy Regulatory Board, India. Radio iodine deposited wires/spheres were placed individually in 100 ml water at room temperature for 48 h, at the end of which the source was removed and the radioactivity in the water was measured by NaI (Tl) well type scintillation counter set for ^{125}I energy.

3.6. Encapsulation and laser welding

Activities of the ^{125}I rods/spheres were measured and sources with activity within ±5 % of the targeted activity (generally 111 MBq/source) were segregated and used for encapsulation. The rods were inserted individually into the titanium capsule followed by placement of a cap over it with the aid of magnifying glass. Capsules were then welded using laser beam using optimised conditions (Fig-4).

FIG. 4. Loading of ^{125}I source core inside the capsules

3.6.1 Welding procedure development

Initially Tungsten Inert Gas (TIG) welding was carried out using a micro torch with the ceramic spacer. Subsequently, a pulsed laser welding system was used for the welding of the capsules. This system consists of a laser head, power supply unit, chiller unit and welding system. As the welding is carried out in pulses, the heat input to the ^{125}I source is reduced. The Nd:YAG laser installed inside the fume-hood in our laboratory is shown in the figure 5. The output of the laser is taken through the ports and it is connected to the welding head using optical fiber.

FIG. 5. Laser welding system

TABLE I. DETAILS OF LASER WELDING SYSTEM

Laser Type	:	Flash lamp excited Nd: YAG
Wavelength	:	1064 nm (IR)
Max. Av. Power	:	50 W
Peak Power	:	8 KW
Output Dia.	:	8 mm
Pulse width	:	2 to10 mSec
Repetition rate	:	up to10 Hz
Aiming Beam	:	< 5 mW, 635 nm red diode laser
Beam Delivery	:	With an optical fibre
Power Supply	:	230 VAC, Single Phase, 16A, 50 Hz

3.6.2 Testing of the welded sources for leak tightness

The inactive welded sources were placed in hot water observed for air bubbles, the absence of which indicated air tightness. The welded samples were also kept in helium chamber at 6 atm. pressure for 22 h and tested by UL-200. Metallography test of the inactive welded capsules were carried out by optical metallography and Scanning Electron Microscopy (SEM).

3.7. Quality control procedures

- The welded capsules were tested for leak proof by the procedures recommended by Atomic Energy Regulatory Board (AERB) India (18). The radio iodine deposited sources in the activity ranges of 1-3mCi were placed individually in 100ml water at 50°C for 5 h, at the end of which the sources were removed and the radioactivity in the water was measured using NaI (Tl) well type scintillation counter set for ^{125}I energy.
- Testing for surface contamination by wipe tests using alcohol immersed cotton wool and checking for radioactive contamination on the cotton wool.
- Accurate measurement of activity by Ion chamber measurements previously calibrated using Onco seed model 6711.
- Source uniformity testing by autoradiography.

3.8. Calibration and dosimetry of sources

The ^{125}I source designed at our center is similar in construction to the Model -6711 seed produced by Medi. Physics Inc.of Nicomed Amersham. The indigenous seed was standardized out using two different well type ionization chambers independently calibrated using Model-6711 seed.

The detailed results have been published [20].The dosimetric parameters of the prepared source were derived in accordance with the recommendations of AAPM Radiation Therapy Committee Task Group (TG)-43 [16]. The parameters were determined experimentally using TLD-100 rods of 1mm (Φ) x 6 mm [l] and perspex phantom of measured density of 1.12g/cc. Details of the dosimetry data generated by Monte Carlo simulation are available in the literature [20].

4. RESULTS

4.1. Electrodeposition

The deposition of activity at various intervals and constant current is given in Table II. 20 μA current for 25-30 min duration resulted in maximum (84%) percentage deposition. The percentage deposition of activity at different currents keeping the time constant is given in Table III. At higher current, the percentage deposition was found to decrease and blackening of silver wire was observed due to oxidation. Table IV shows that the percentage deposition of ^{125}I that did not vary with radioiodine concentration in the bath. The leach test showed a low percentage of leachability (<0.05%) indicating firm deposition of radioactivity.

TABLE II. EFFECT OF TIME ON THE ELECTRODEPOSITION OF ^{125}I ON SILVER WIRE

Time period of electrodeposition (min)	Amount of ^{125}I deposited on wire μCi(SD)	Percentage adsorption(±SD)
5	238.33(6.23)	47.66(1.24)
10	281.33(8.65)	56.33(1.74)
15	346.66(6.23)	75.40(1.60)
20	377.00(8.04)	75.40(1.60)
25	420.66(8.99)	84.00(1.63)
30	410.00(7.81)	82.12(1.34)

(Activity in the BATH=500 μCi, CURRENT=20μA, *n*=3)

TABLE III. EFFECT OF CURRENT ON THE ELECTRODEPOSITION OF ^{125}I ON SILVER WIRE

Current passed for deposition(μA)	Amount of ^{125}I deposited on wire μCi(SD)	Percentage adsorption(±SD)
10	385.66(9.97)	77.13(1.99)
20	413.33(6.23)	82.66(1.24)
30	395.00(8.165)	79.00(1.63)
40	363.33(8.49)	72.66(2.30)
50	350.00(7.81)	70.00(1.40)

(Activity in the Bath =500 μCi, TIME=25 min, *n*=3.)

TABLE IV. EFFECT OF ^{125}I CONCENTRATION ON ELECTRODEPOSITION

Initial activity in the bath(μCi)	Amount of ^{125}I deposited on wire μCi(SD)	Percentage adsorption(±SD)
200	164.66(2.49)	82.33(1.25)
400	334.66(6.79)	83.66(1.74)
800	395.00(8.165)	83.54(1.47)
1600	363.33(8.49)	84.33(0.94)
3200	350.00(7.81)	84.00(0.81)

(Current: 20 μA, time: 25 min, *n*=3.)

4.2. Physical adsorption

The percentage adsorption of ^{125}I on the silver wires was low (~ 20 %), when the adsorption was carried out at room temperature and did not significantly improve even at higher temperatures (40-60^{o}C). Moreover these results were not consistent and reproducible. The percentage adsorption was irreproducible and low, irrespective of the volume of radioactive iodine solution taken for adsorption. The adsorbed activity that leached out from the silver wires was high (~4%) which is above the permissible limits.

4.3. Physico-chemical adsorption

Maximum adsorption of radioactive iodine could be obtained on the Palladous chloride treated silver wires and the percentage adsorption was inversely proportional to the volume of radioiodine solution. Though there was noticeable adsorption of activity (30-40%) at room temperature, the percentage adsorption increased with increase in temperature up to 60°C and remained nearly constant up to 70°C (Fig. 6). The time duration of heating also played a significant role on the percentage adsorption. The adsorption increased with increased duration of heating and reached a maximum (90%) at 6h. The carrier addition was critical in the adsorption of ^{125}I on the Pd treated silver wires. The percentage adsorption was 50% without the addition of the carrier which increased to 90% at 5-10µg of carrier (Fig.7).The ^{125}I coating was uniform as was seen by auto radiography and showed negligible leachability (<0.05%).

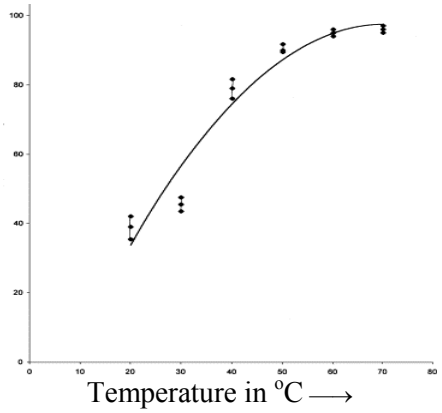


FIG. 6. Effect of temperature on the percentage adsorption on Pd coated silver wires

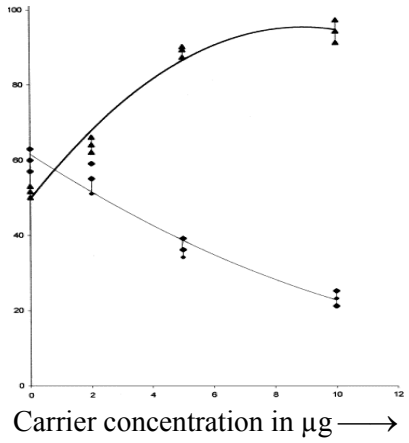
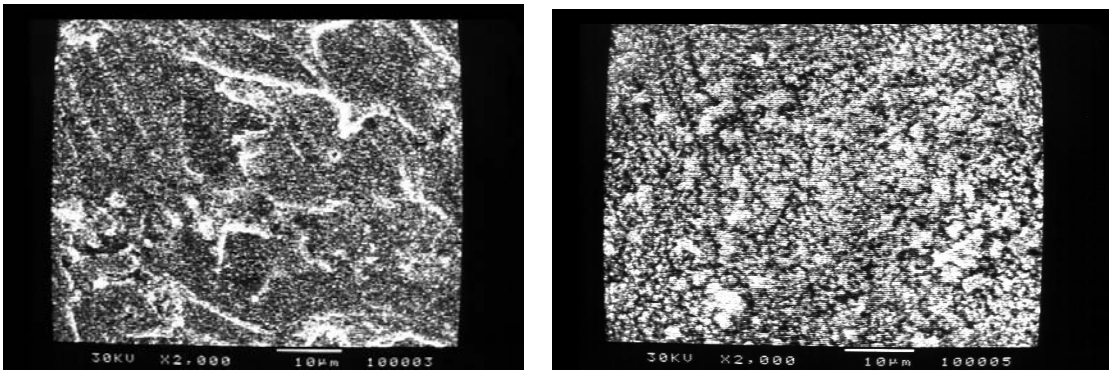


FIG. 7. Effect of carrier concentration on percentage adsorption of Pd coated and bare silver wires

The Scanning Electron Microscopy photographs of uncoated and coated Silver Rod are shown in Fig.8. It is observed from the result that metallic palladium is distributed uniformly through out the surface of the silver rod.



(a)Uncoated Silver Rod (b) Palladium Coated Silver Rod

FIG. 8. Scanning Electron Microscopy photograph of Silver Rod

4.4. ^{125}I coated by physicochemical adsorption on alumina spheres

Fig.9 shows scanning electron microscopy of a typical alumina microsphere prepared by internal gelation process. The 50 times magnified picture indicates the porous nature of the micro sphere.

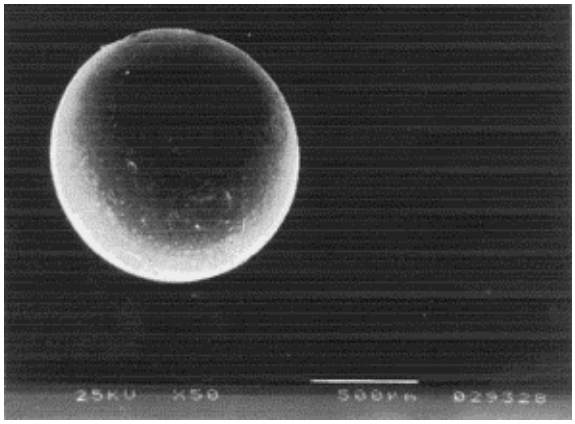


FIG. 9. Electron micrograph of a microsphere

The rate of adsorption of radioiodine as iodate (IO_3^-) on the microspheres is shown in Fig.10. It was apparent that the adsorption of iodine as iodate progressively increased with time and reached a maximum of 95% at 4 h and remained nearly constant up to 5h.

It was also observed that for quantitative adsorption, the volume of radioactive solution should be kept at minimum possible. The phenomenon was similar to soaking and at higher concentrations of radioiodine better adsorbtion was seen as shown in Table V.

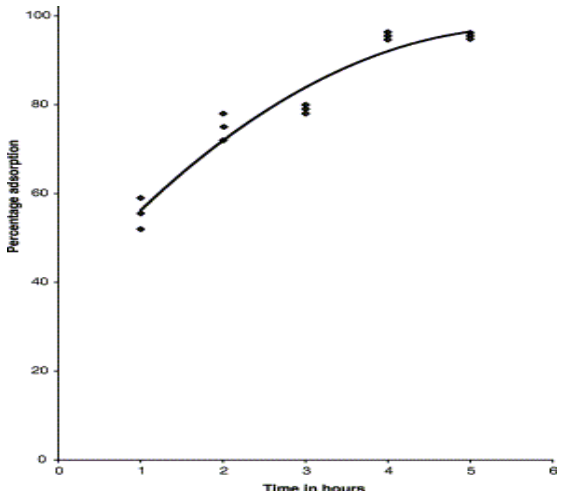


FIG. 10. Adsorption of ^{125}I as IO_3^- on alumina microspheres vs. time.

TABLE V. EFFECT OF ^{125}I CONCENTRATION ON ADSORPTION

Initial activity in the bath (μCi)	Amount of ^{125}I deposited on sphere in μCi (SD)	Percentage adsorption (±SD)
200	190.00(4.08)	95.00(2.04)
400	393.00(2.16)	98.00(0.54)
600	578.00(5.88)	96.33(0.98)
800	784.00(7.11)	98.00(0.89)
1000	979.00(6.16)	97.90(0.61)
1500	1472.33(5.88)	98.13(0.39)

4.5. ^{125}I coated by physicochemical adsorption on treated silver spheres

The quantitative adsorption of ^{125}I could be obtained on 0.5 mm (ϕ) palladium coated silver beads. The percentage adsorption was found to increase with the increasing amount of carrier and attained a maximum value of ~ 80%, using ~1μg of carrier [Fig.11]. With further increase in carrier amount, a decrease in adsorption was noticed.

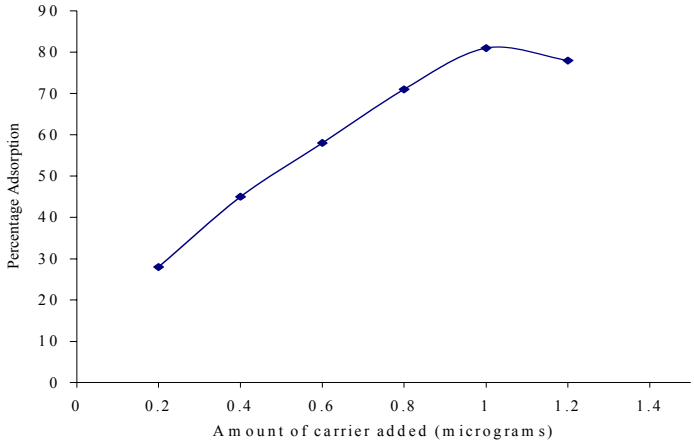


FIG. 11. Effect of carrier amount on percentage of adsorption

The adsorption was also found to increase with the increase of temperature upto ~60-70ºC and this temperature was found to be optimal for maximum adsorption (Fig. 12).

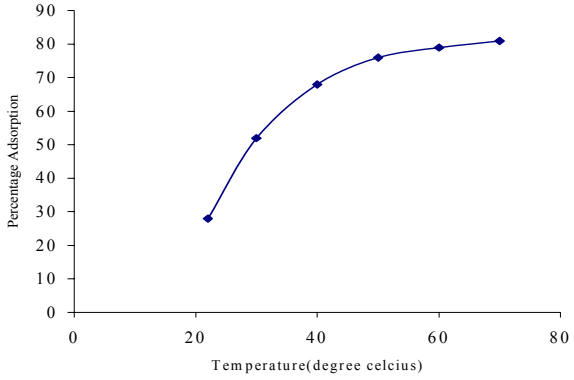


FIG. 12. Effect of reaction temperature on percentage of adsorption.

The radioactive beads could be prepared with extremely good reproducibility (CV ~ 8-10%) and three batches of beads, each having ~26 MBq (0.7 mCi) of ^{125}I could be prepared. The variation in the values of source activity measured at two different places i.e. RPhD and RP&AD was found to be within ± 5%. The leachability of the ^{125}I-beads was found to be ~0.007%, which is less than the AERB limit of 0.01% of the source activity.

4.6. Uniformity of ^{125}I activity

The results of autoradiography showed that the distribution of activity on the Pd coated silver wire source was uniform with a coefficient variation of <7%.

Table VI depicts the optical density distribution of 125I sources randomly selected from each batch.

TABLE VI. OPTICAL DENSITY DISTRIBUTION OF ^{125}I SOURCES FROM EACH BATCH.

Direction	Batch Number					
	1.6	1.3	1.5	1.7	1	1.9
	1.4	1.4	1.5	1.6	1	2
	1.5	1.3	1.4	1.7	1.1	1.9
	1.6	1.2	1.6	1.7	1.1	1.8
	1.7	1.5	1.5	1.5	1.1	2.2
	1.5	1.4	1.5	1.6	1.1	2
	1.6	1.4	1.4	1.5	1	2
	1.5	1.4	1.5	1.5	1.1	2
Average	1.55	1.36	1.48	1.6	1.06	1.96
RSD	5.97	6.72	4.31	5.78	4.48	4.67

4.7. Leachability of the ^{125}I adsorbed activity

The protocol followed for testing the leachability of the final sources was as prescribed by Atomic Energy Regulatory Board, India (AERB), described earlier. Table VII summarises the results of the leachability studies.

TABLE VII. LEACHABILITY OF ^{125}I SOURCES

Details of the sources	Bare silver wires	Palladium coated silver wires	Electro-deposited silver wires	Alumina spheres	Palladium coated silver spheres
Range of activity(MBq)	44-112	111-148	111-148	35-40	15-20
% Activity leached out	~ 4	~ 0.01	~ 0.05	~4.0	~0.01
No of sources studied	50	115	60	40	50

The leachable activity of palladium coated silver wires were far lower (0.01%) than that of the electrodeposited silver wires, although both are within the permissible limit. In the case of spherical sources, the leachability on alumina spheres was much higher (~4%) than that of palladium coated silver spheres (0.01%). This indicates that pre treatment of the silver source with palladium facilitates firm chemical bonding of iodine on the surface that does not leach out.

4.8. Encapsulation

A large number of capsules were welded varying the different parameters. Fig.13 shows the cross-sectional view of titanium encapsulated wire source and microspheres.

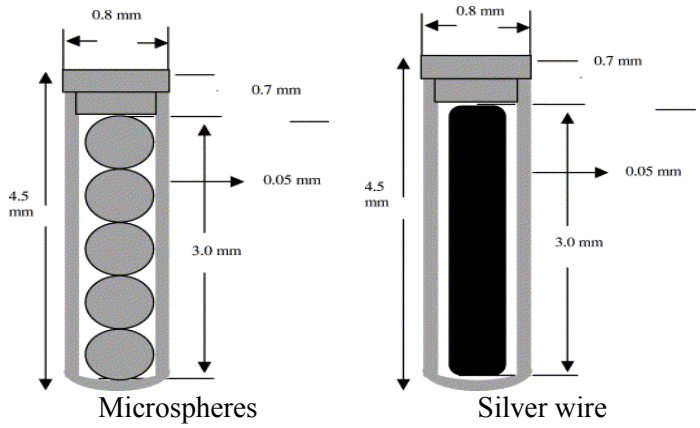
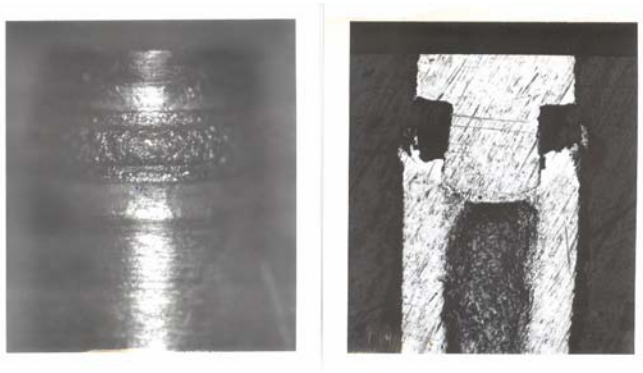


FIG. 13. Cross sectional view of encapsulated I-125 source.

Tungsten Inert Gas (TIG) welding is the most widely used process for welding titanium. In this process the hot zone being welded is shielded from the atmosphere to prevent contamination with oxygen and avoid degradation of the ductility of the welded portion. Therefore, initially we have carried out the welding of the capsule using this technique. However, since the capsules were of only 50 micron thick, it was observed that on TIG welding the titanium melted. The welded portion had perforations while the molten metal flowed along the sides. It was also observed that during the welding, ^{125}I evaporated and settled in the welded porous area, resulting in leakage of activity.

To overcome this problem, an Nd:YAG pulsed lasers laser welding system was procured and used for the welding of the capsules. The depth of the weld and the thickness of section that can be welded are primarily determined via the laser power. The welding parameters such as energy, frequency and pulse duration were optimized by welding inactive capsules to give leak proof welding.

All these welded capsules were analyzed metallographically. Based on the results, the best parameters were identified. Another set of capsules were welded with the identified parameters and metallography tests were carried out to reconfirm the parameters. It was found that if the wall thicknesses of capsule and cap are nearly equal and the beam is focused at the centre of the joint, good quality leak-proof welding can be achieved.



(a) **(b)**

FIG. 14. Optical metallograph of Welded capsules (a) Outer Surface (b) Inner cross-section

The metallography test of welded capsules was carried out by optical metallography (Fig.14) and Scanning Electron Microscopy (Fig. 15). The penetration depth in the samples was found to be ~ 2-3 times the wall thickness of the capsules. The welded samples showed high integrity and superior metallurgical quality.

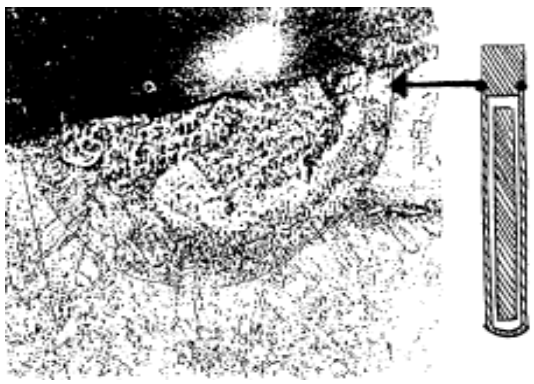


FIG. 15. SEM of welded capsule

4.9. Dosimetry

The mean value of the air kerma strength (±2%) measured using the two types of chambers was used for establishing the dosimetric parameters of the new source. The dosimetric studies revealed that the dose rate constant (Λ) of 0.95 ± 0.06 cGy $h^{-1}U^{-1}$ was in good agreement with the value quoted for 6711 type seed i.e. 0.97 ±2% cGy $h^{-1}U^{-1}$ [20]. The major uncertainties in the measurement of dose using TLD rods include uncertainty in source calibration (~5%), uncertainty in response of TLD among the rods (~3%), energy correction relative to ^{60}Co (~2%), positional uncertainty (~ 1%) and dose conversion from perspex to water (~1 %). The overall uncertainty in the entire measurement works out to be ~6.3%**.** The measured dose rate constant of 0.95 ± 0.06 cGy h^{-1} U^{-1} for the new seed is in good agreement with the Monte Carlo calculated value of 0.97 for similar model seed (type 6711). The radial dose function g(r), at radial distance of 0.5, 1.0, 2.0, 2.5 and 3.0 cm (Table VIII) are also in agreement with the values quoted for 6711 type seed, within the uncertainty of measurement (~6%).

The mean value of measured and reported values of dose rate constant (Λ= 0.96) was considered useful for the clinical dosimetry of the source. Figure 16 shows the experimental arrangement for dosimetry.

TABLE VIII. RADIAL DOSE FUNCTION; G(R)

Distance from source (cm)	Model 6711 Seed (Bret.H.Heintz et.al, 2001).	Measured value of source under consideration
0.5	1.04	1.10
1.0	1.00	1.0
2.0	0.832	0.815
2.5	0.731	0.744
3.0	0.632	0.671

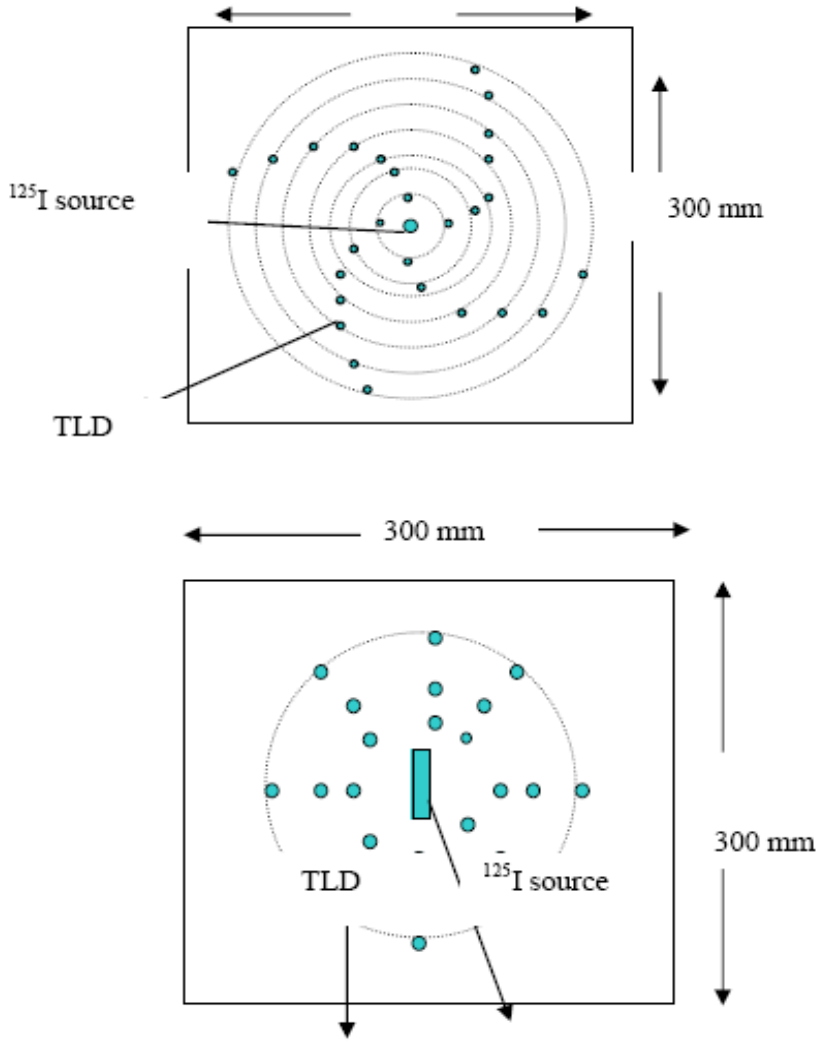


FIG. 16. Experimental arrangement for dosimetry

4.10. Quality assessment

The results of the quality assessment of the welded capsules are given in Table IX. The average activity per source was 2 ± 0.19 (SD) mCi with a coefficient variation of 10 % which is within prescribed limits. The surface contamination of all the prepared sources was less than 0.5 nCi which is well within the prescribed limits. The total release of activity from all the welded capsules in the immersion test at 50^{o}C for 5 h was less than 5 nCi which is well below the permissible limits.

TABLE IX. QUALITY ASSESSMENT OF WELDED CAPSULES

Source Code No.	Activity (mCi) as on 25/08/03	Surface Contamination (nCi)	Total release in 50°C/ 5 h immersion test (nCi)
B-2/1	2.01	0.02	3.81
B-2/2	2.11	0.01	4.8
B-2/3	2.20	0.02	3.4
B-2/4	1.90	0.02	3.6
B-2/5	2.16	0.13	4.2
B-2/6	2.13	0.02	3.6
B-2/7	1.92	0.06	4.6
B-2/8	2.22	0.001	2.76
B-2/9	1.83	0.04	4.1
B-2/10	1.78	0.04	3.9
B-2/11	1.76	0.04	4.6
B-2/12	1.94	0.03	4.1
B-2/13	1.76	0.03	1.2
B-2/14	1.60	0.03	1.6
B-2/15	1.65	0.09	3.8
B-2/16	1.76	0.01	2.8
B-2/17	1.83	0.01	3.9
B-2/18	1.72	0.06	3.7
B-2/19	1.61	0.36	4.7
B-2/20	1.90	0.03	4.2
B-2/21	1.81	0.02	3.9
B-2/22	2.22	0.001	4.1
B-2/22	1.99	0.002	0.8

Average activity per source 2 ± 0.19 (SD mCi), % CV = 10

5. DISCUSSION

The electro deposition method of preparing ^{125}I source core is a straight forward procedure and used by many commercial manufacturers of these sources. Quantitative firm and uniform deposition could be obtained by the optimized parameters, with no leaching of activity. Increasing the current to reduce the time of deposition resulted in lower deposition of activity; this might have been due to the oxidation of silver in the aqueous medium. However this method, at our hands had some practical disadvantages. To precisely cut the radioactive coated wire manually to the required miniature source dimensions (~ 3.5 mm length) so as to fit in to the ISO specified dimensions of the Titanium capsules (4.5 mm length) would be labour intensive, require high skill and would be exposing personnel to long time exposure to radiation. Additionally, there could be wastage of deposited activity. The optimized time of 25 min per wire piece would necessitate multiple deposition processes for preparation of large number of sources. This perhaps could be done by use of the same bath for either sequential deposition or for parallel deposition on silver wire cathodes. But this can be resorted to only if uniform and reproducible deposition can be ascertained.

Thus the electro deposition method was found to be attractive only if automated where radioactive iodine is electrodeposited on a long piece of silver wire of the required diameter and subsequently precision cut in to the required dimensions by remotely operated electro mechanical cutting devices.

Physical adsorption of ^{125}I activity on the silver wires under the experimental conditions studied was not quantitative and reproducible. The variation in percentage adsorption from 20% to a maximum of 60% under different experimental conditions seemed to be dependent on the amount of carrier iodide due to the limited adsorption sites on the surface for forming silver radioiodide. The

desorption or leachability of these sources is perhaps due to the solubility of silver iodide which is higher than that of palladous iodide.

Physical adsorption of ^{125}I activity on the $PdCl_2$ treated silver wire were more suitable for adsorption of ^{125}I than plain silver wires in terms of quantitative adsorption and non-leachability of ^{125}I activity. The mild experimental conditions of adsorption at neutral to alkaline pH facilitates the safe handling of high amounts of radioactivity for the preparation of high intensity therapeutic sources without the release of air activity. The stability of ^{125}I on Pd coated silver wire is due to the formation of insoluble Palladous Iodide on the surface of the wire and accounts for low leachability.

The physicochemical adsorption of radioiodine on alumina microspheres is easy and also less expensive. Quantitative adsorption of activity on the spheres is possible by using radioactive iodine in iodate (IO^{-}_{3}) form. However, it was found that the percentage leachability of radioactivity from the spheres was slightly more than desirable. More investigations were needed to overcome this problem. A large batch of alumina spheres of appropriate size could be easily prepared and used for quantitative adsorption of radioiodine as iodate (IO_3^{-}). One of the methods to reduce leaching is by having an organic coating after the adsorption of ^{125}I. This work is in progress.

The physicochemical adsorption of Pd-silver microspheres is easy and ^{125}I-beads could be prepared in a nonleachable form. The radioactive sources upto ~111MBq (3 mCi) activity can be prepared by arranging five individual beads in a well-arranged geometry. As the source design of tiny radioactive sources plays a vital role in the dosimetric aspects, the newly developed spherical ^{125}I-beads on account of relatively higher dose rate constant may reduce the total treatment time and have potential to replace the presently used rod type ^{125}I-sources in eye and prostate brachytherapy. Titanium was chosen as the capsule material because of the following properties:

- high strength to weight ratio;
- corrosion resistance;
- mechanical properties at elevated temperatures;
- amenable to easy welding.

Titanium is a unique material, as strong as steel but half its weight with excellent corrosion resistance. The high strength, low weight and outstanding corrosion resistance possessed by titanium and titanium alloys have led us to select as capsule material.

The welding of capsules containing radioactive sources requires that the following conditions are met:

- The welding has to be carried out in a well ventilated fume hood.
- The process of welding should be feasible in a remotely operated system.
- The welding should not change the geometry of the capsules.
- Loss of activity during the welding should be minimum(minimum heat input to the active source)
- Efficiency of welding should be such that there should not be leakage of activity after welding.(Welding should cause less porosity)

Although TIG process is the most common method for welding titanium materials, this is not suited to our type of application as mentioned earlier. The molten titanium was observed to flow down in TIG welding and the weld was observed to be porous.

The most likely cause of porosity is the trapping of gas bubbles between dendrites during solidification and presence of hydrogen from moisture in the arc environment. As the leakage of radioactivity from the welded capsules are significantly higher than the prescribe limit, alternative method of welding needed to be investigated to overcome this problem.

To overcome these problems, a pulsed laser welding system was used for the encapsulation of capsules. Major advantages of laser beam welding are low welding stresses, low risk of distortion, creation of minimal heat affected zone with minimal ^{125}I contamination and capable of welding of varying mass that allows hermetic sealing of Ti capsules. Nd:YAG pulsed lasers have the ability to weld hard materials like Ti and produce an aesthetical weld with high depth/width ratio free from any weld buildup that eliminates many secondary operations such as grinding or honing. It has also high welding speed, good reproducibility, flexibility and the process can be easily remotised and automated. Due to the extreme reactivity of titanium metal, it is essential to shield the molten pool and the hot metal from contact with air. Nitrogen is used as inert gas protection as it is cost effective.

These capsules were then used for carrying out clinical trials at Sankara Netralaya, Chennai, a leading eye care hospital in India. After approval from the AERB, 5 patients were treated using the seeds prepared by us. Out of these five cases, two cases are of retinoblastoma and three cases are of malignant melanoma. The results are as follows:

- Out of three cases retinoblastoma, two cases did well ultimately while one case showed too rapid a regression followed by severe recurrence needing removal of eye.
- Both the cases of malignant melanoma were doing well. But could not be followed up after 2 months.
- One case of retinoblastoma showed remarkable improvement. This patient was treated after unsuccessful External Beam Radiation Therapy, which is highly gratifying.

Although the number is very low to conclude the efficacy of the treatment, it is highly encouraging to know that brachytherapy using ^{125}I sources has been effective in certain category of patients and enucleation of eye could be prevented. It is also note worthy that often external beam radiotherapy fails in treatment of ocular tumours which respond well to the brachytherapy treatment.

6. CONCLUSIONS

6.1. Capabilities built

- Preparation of ^{125}I sources based on Pd coated Ag matrix in high yields, reproducibility and large scale.
- Laser welding of the sources encapsulated in titanium capsules.
- The sources could be deployed for treatment of ocular cancers with the help of an eye care hospital. The sources are now awaiting approval for regular production and supply.

6.2. Impact

Since the availability of these sources in the country, the number of hospitals asking these radioactive seeds for undertaking plaque therapy has increased. It is still too early to judge the qualitative impact of this technology.

6.3. Future plans

Thus far a technique for the production of iodine-125 brachytherapy sources has been developed. The next challenge will be to develop more complex sources containing higher activity of iodine-125 (~500 mCi) for their possible application in Lexiscopy and bone desitometry.

6.4. Partnerships

Several divisions of BARC (Laser processing and advanced welding section, Centre for design and manufacture, Isotope applications, Radiometallurgy, Post irradiation examination, Radiological physics and Advisory) as well as external agencies like Atomic Energy Regulatory board and Hindustan Machine Tools Ltd. have contributed towards this development.

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DEVELOPMENT OF ^{125}I SEED SOURCE FOR TREATMENT OF EYE AND PROSTATE CANCER

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Abstract

The adsorption of ^{125}I on silver bits coated with palladium was studied. The adsorbed ^{125}I on Pd coated silver wires could be used as matrix for the preparation of interstitial source. Effective parameters on adsorption of ^{125}I such as temperature, time and carrier volume and concentration were investigated. These experiences showed quantitative and consistent uptake (83%) of ^{125}I and exhibited low leach ability (0.06%). The method is under optimization to obtain higher adsorption of ^{125}I on the palladium treated silver wires. A laser welding machine used for welding of titanium capsules and welding parameters were investigated. Dosimeteric parameters of solid core ^{125}I were determined by radiochromic MD-55-2 GAFCHEROMIC film dosimetery. The results of the film dosimetery indicate that nonuniformity of activity around the core is about 8%, also the dose rate at distance of 1cm for active core was 1.133 cGy/hr/mCi. Iso-dose curves around of ^{125}I core have been plotted for 90, 70, 50, 30 and 10 percents using simple image processing software. The results indicated that uniformity of coated activity was good. Production of ^{192}Ir for HDR machine was carried out. 10 Ci ^{192}Ir source with the size of 4.6 mm outer diameter and 6.5 mm length connected to flexible cable by TIG welding was fabricated and sent to hospital for further investigation.

PART A: ^{125}I SEED SOURCE PRODUCTION.

1. INTRODUCTION

^{125}I seeds are indicated for interstitial treatment of tumors which have the following characteristics: localized, slow growth rate and low to moderate radio sensitivity. They are also indicated for the treatment of recurrent tumors and residual tumors following a course of external radiation therapy. ^{125}I seeds may be used to treat superficial, intra abdominal and intrathoracic tumors. Tumors of the head, neck, lung, pancreas, eye and prostate are commonly treated. Interstitial brachytherapy using ^{125}I sources for treatment cancer and as permanent implants for tumors have a recognized place in the radiation therapy modality.

However, to produce an effective source, and to meet the requirements for its immediate use, seeds sealed in a titanium capsule by laser welding. ^{125}I seeds emit X rays of 27.4 and 35.5 keV. The dose distribution around each individual seed is not isotropic. However, dosimetric aspects of these sources have been reported from time to time. There are several methods for production of core for ^{125}I sources. Adsorption of ^{125}I on silver wire as matrix for brachytherapy sources [1], ^{125}I deposition on silver wire by electrochemical techniques [1,2] and adsorption on alumina microspheres. After trying different method we were successful in making the core of ^{125}I by adsorption of ^{125}I on silver wire. The studies include selection of laser welding machine for welding of titanium capsules of ISO specific dimension, activity assessment of source core in term of activity and uniform adsorption of activity along the core of ^{125}I [4].

2. MATERIAL

^{125}I as sodium iodide in dilute sodium hydroxide solution was obtained from MDS Nordions. Silver wire of 0.5mm (ø) with p.t. 99.9% was obtained from Chemical Sciences Co. Tehran and cut to 3 mm length mechanically; palladous chloride was provided by Merck-Schuchardt, 8011 Hohenbrunn bei Munchen. Kodak film grade-D7 was used for autoradiograghy.Titanium tube (ASTM F67) of 0.0305/0.0325 inch OD: 0.0015 /0.0025 inch. Wall length: 0.175/0.179 inches tubing/ seamless ½ hare temper titanium grade 2 was obtained from KERI through CRP cooperation.

Titanium foil of 0.05 mm thickness and rod of 0.81 mm diameter were obtained from Alfa Company for cap manufacturing. Welding system, Nd: YAG laser beam was used to weld capsules of 0.8 mm (OD) and 3-6 mm (L) (pulse type). The technical parameters were: wavelength 1064 nm, maximum energy/pulse 60 J, maximum peak power 5 kW, average power 15 W., maximum repetition rate 10 Hz. For cooling system heat exchanger and chiller with 3 kW cooling capacity were applied. The viewing system was fiber-based with connection to the monitor and the CCD camera for getting the image of the fiber optics. The job handling system has rotating assembly, which can hold cylindrical work pieces of dimension 0.8 mm (OD) and 3-6 mm (L). The rotation of spindle was controlled by an electronic card which acted on the stepping motor. The speed variation of the job was from 1 up to 100 revolutions per minute with preset facility for selecting and reading the speed value. A gas-shielding arrangement with a solenoid valve was installed to protect the titanium during welding. An ion chamber was used for the dosimetery of core activity.

3. METHODES

3.1. ^{125}I core production

In this work production of ^{125}I core with use of chemical adsorption technique was studied and influence of some effective parameters such as method of silver wire coating with palladium, temperature, carrier volume and concentration and time on ^{125}I adsorption were investigated.

3.1.1. Effect of silver wire coating method

A comparative evaluation of palladium optimal composition coated silver wire and palladium single coated silver wire on the adsorption of ^{125}I was made. While the adsorption of palladium optimal composition coated silver wires showed low, inconsistent uptake (about 21%) of ^{125}I, the palladium single coated silver wires showed quantitative and consistent uptake of ^{125}I (about 83%).

3.1.2. Effect of time, temperature and carrier volume and concentration on the ^{125}I adsorption (palladium single coated silver wire)

For investigating influence of operational variable such as time, temperature and carrier volume and concentration on the ^{125}I adsorption we arranged a set of experiments which some of theme mentioned here.

A volume of 40 micro litter containing about 4 mCi ^{125}I and 20 micro litter of KI 0.03 M as carrier was maintained. The adsorption was carried out at 80 degree centigrade from 30 to 360 minute. The effect of time on the adsorption of ^{125}I on the palladium single coated silver wire was studied.

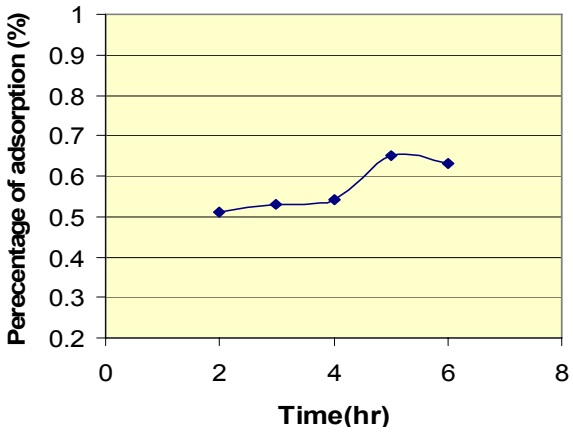

FIG. 1. Effect of time on adsorption of ^{125}I at 80 degree centigrade

A volume of 40 micro litter containing 4 mCi (148 MBq) of ^{125}I and 10 micro litter KI 0.03 M as carrier was maintained .The adsorption was carried in 60 degree centigrade. The effect of time on the adsorption of ^{125}I on the palladium single coated silver wire was studied. The time of adsorption was varied from 30 to 540 minutes (Fig.2).

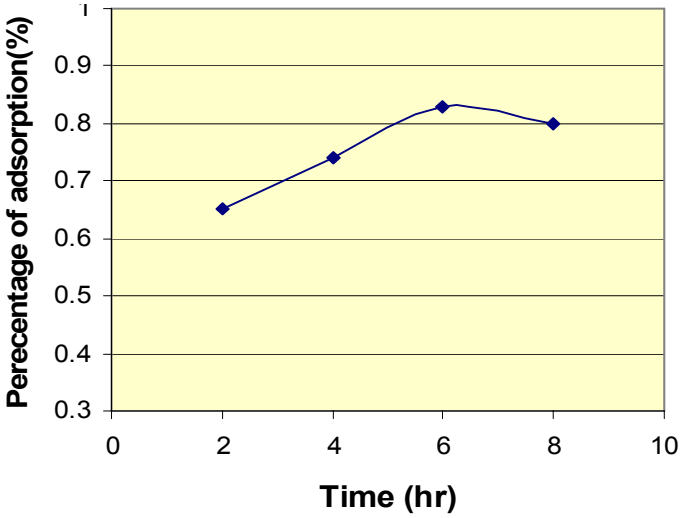

FIG. 2. Effect of time on adsorption of ^{125}I at 60 degree centigrade

The above experiments were repeated using KI as a carrier ranging from 1 to 30 µg in a volume of 10 µl keeping the reaction volume as 50 µl and keeping the other optimized parameter constant. The effect of carrier concentration on ^{125}I adsorption is shown in Fig.3.

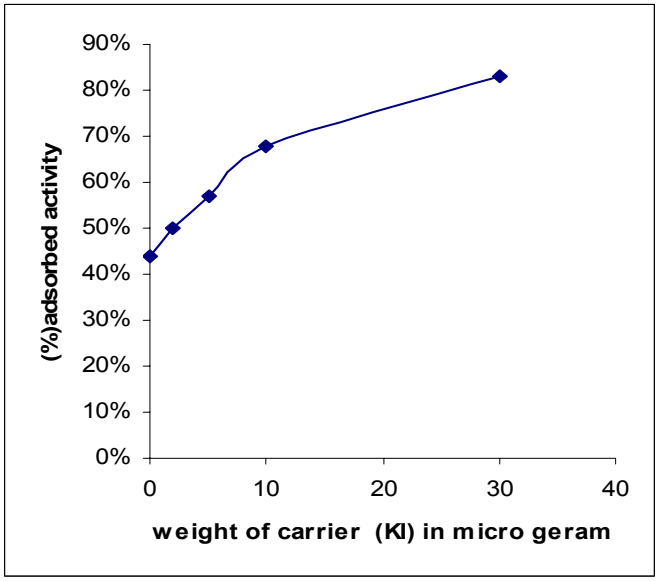

FIG. 3. Effect of carrier concentration on adsorption of ^{125}I at 60 degree centigrade

After selection of above parameters five ^{125}I cores was produced simultaneously. The carrier was KI and the initial activity for production of five sources was 2.5 mCi (92.5 MBq).The core activities produced were 370, 330, 300, 328, 290 µCi respectively. Investigation on using of some other materials as a carrier in producing ^{125}I core has to be carried out in the near future.

3.2. Determination of uniformity activity of adsorbed ^{125}I on the silver wire

For determination of uniformity activity of ^{125}I adsorbed silver wire two different methods were used as bellow:

A circle disc of 4.4 cm diameter and thickness of 1.4 cm with eight equidistance tunnel of uniform aperture at an angle of 45 degree were drilled through central hole. Autography of a single source was taken simultaneously from eight equidistance directions placing the source in centre and film wrapped around the disc. The result of density is shown in the (Fig.4).

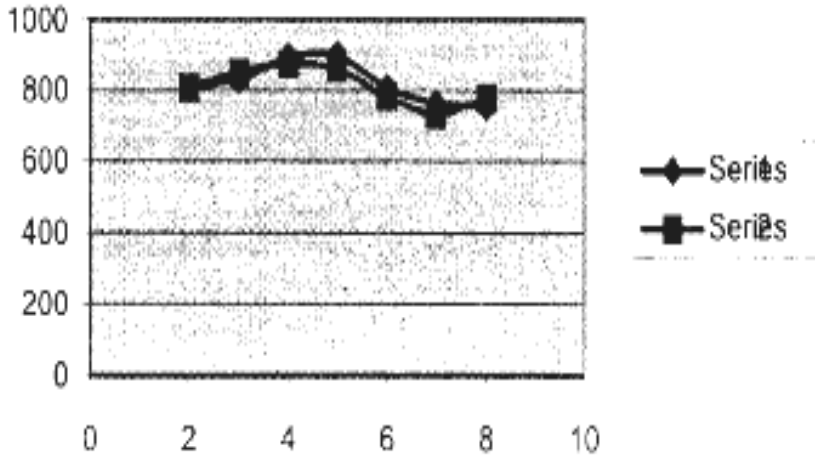

FIG. 4. Investigation of linear adsorption of ^{125}I along the core

To determine nonuniformity of activity and dose distribution of produced core; dosimetry was performed using MD-55-2 GAFCHROMIC film. Nonuniformity of activity on transverse axis can be determined by measuring axial dose distribution in a pelexiglass phantom. After irradiation of the films, their responses were measured using a spectrophotometer in wavelength of 660 nm. For dosimetry we used MD-55-2 film with size of 4 x 4.5 cm^2. Irradiation time of film was 60 hr. After irradiation the MD-55-2 film was cut into 9 segments with size of 1.3 x 1.5 cm^2. In figure 5 the schematic of the cut films with label no. 1 to 9 was shown. Amount of net O.D. with respect to each film segments were shown in table I.

9	3	8
5	1l 1c 1r	4
7	2	6

FIG. 5. Schematic of the cut film and considered position arrangement on the film. The position 1 is a source palace

The MD-55-2 film segment respect to core position with label no. 1 was shown in (*Fig. 6)*.

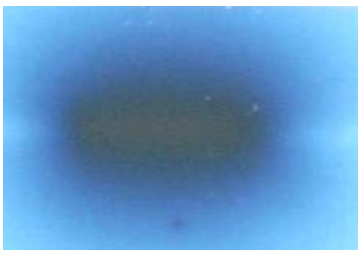

FIG. 6. Schematic of MD-55 film with respect to core position (1), Y axis is in the length of seed

Data of this film was included 1c (center of core), 1l (left section of core) and 1r (right section of core) that were shown in table I.

TABLE I. RESULT OF FILM DOSIMETRY, Y-AXIS IS ALONG OF CORE.

Pos. no.	1c	1l	1r	2	3	4	5	6	7	8	9
X position(mm)	0	0	0	12.5	-12.5	0	0	12.5	12.5	-12.5	-12.5
Y position (mm)	0	-2	2	0	0	17	-13	17	-13	17	-13
Net OD.	1.215	0.675	0.575	0.125	0.115	0.095	0.16	0.063	0.096	0.095	0.065
Dose (Gy) , ^{60}Co	54.9	36.1	31.67	7.967	7.37	6.11	10.8	4.22	3.95	3.93	4.20
Dose (Gy), ^{125}I	91.45	60.17	52.78	13.28	12.28	10.1	18	7.03	6.58	6.55	7
Dose rate (cGy/hr/mCi)	12.20	8. 03	7. 04	1.77	1. 63	1.36	2.4	0. 94	0. 88	0. 88	0.93

The measured O.D. was converted to dose rate, by calibrating samples of these films using the ^{60}Co gamma rays, (Fig.7). Calibration of the MD-55-2 films is obtained using a ^{60}Co Teletherapy unit.

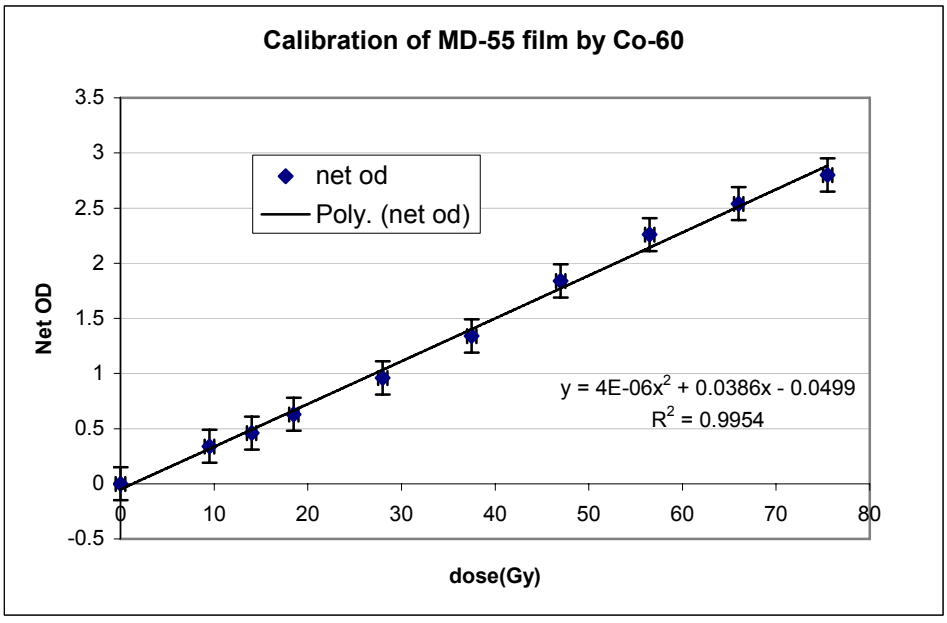

FIG. 7. Calibration of the MD-55-2 films

Iso-dose curves around ^{125}I have been plotted for 90, 70, 50, 30 and 10 percents using simple image processing software, and have been analyzed with a qualitative color image, (Fig.8 and Fig.9).

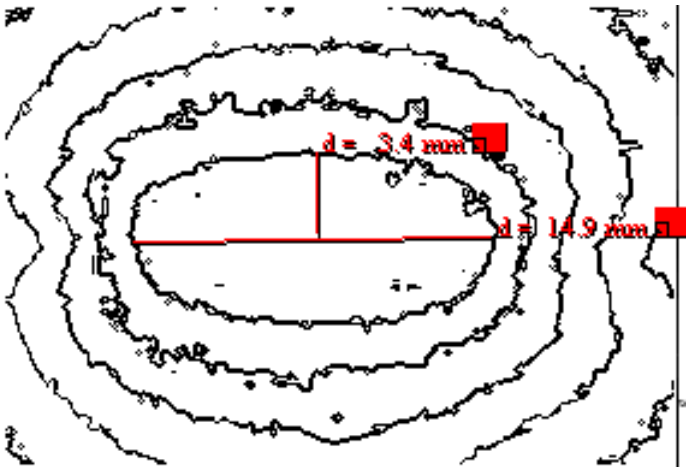

FIG. 8. Iso-dose curves around ^{125}I core have been plotted for 90, 70, 50, 30 and 10 percents using simple image processing software.

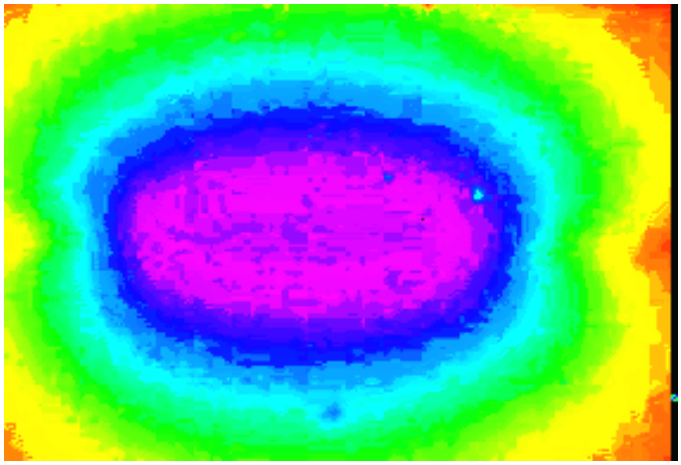

FIG. 9. Iso-dose curves around ^{125}I core has been showed with a qualitative colour image using simple image processing software

3.3. Laser welding

Development of the technique/device for assembling and sealing of ^{125}I source with precise positioning, source core encapsulation and welding technique was considered. Due to oxidization nature of titanium the laser welding for titanium capsule was preferred to other welding methods such as plasma and TIG welding. A new Nd-YAG laser system was prepared exclusively for this work. The welding parameters such as energy and pulse duration were systemically controlled to obtain good uniform welding. The welding was satisfactory by visual test and microscopic pictures (Figs. 10, 11, 12). The optimal welding was performed with energy of 7-8 J/pulse, for 4 msec pulse duration, with 1 HZ frequency and 80 pulses per one revolution .The nitrogen gas with flow of 18 liter per minute was used as a shielding gas.

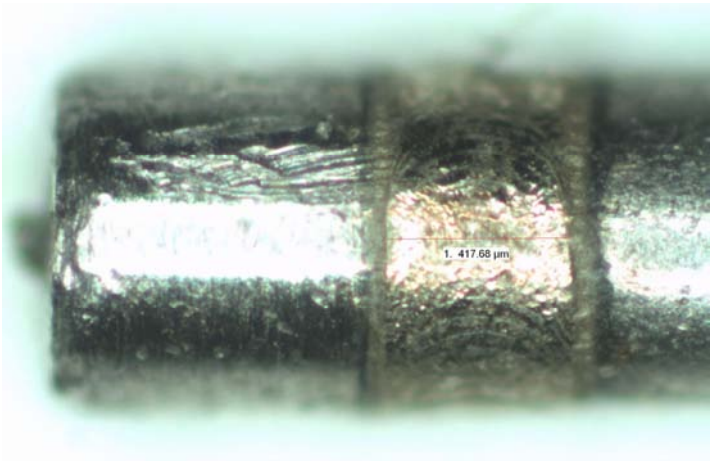

FIG. 10. Laser welding of titanium cap and tube.

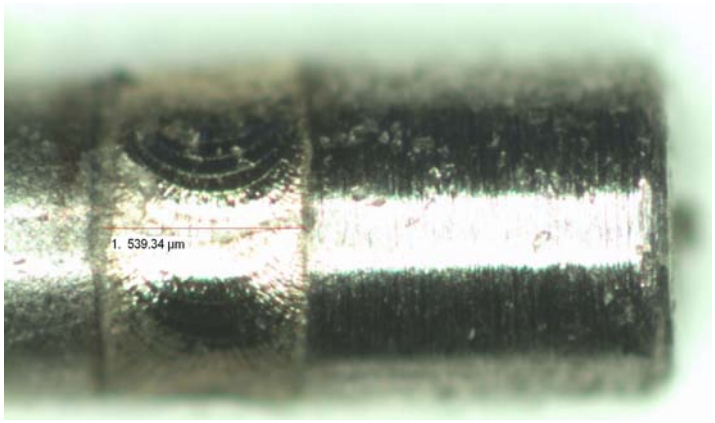

FIG. 11. Laser welding of titanium cap and tube.

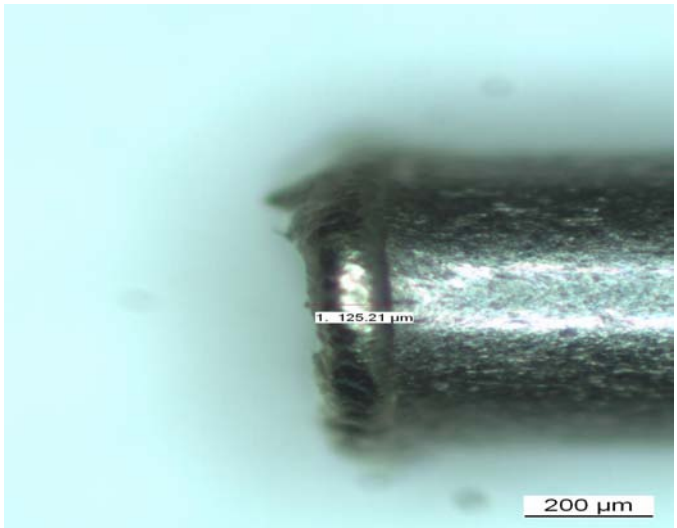

FIG. 12. Laser welding of titanium tube with 0.05 mm Ti cap.

3.4. Leak test

The test for titanium sealed capsule was performed by liquid nitrogen leak test as follows. The empty seed was immersed for about 5 to 10 minutes in 250 CC backer filled with liquid nitrogen. Then the sealed capsule removed from the liquid nitrogen bath and immediately placed in a bath of water at room temperature. Any liquid nitrogen in the capsule will be rapidly vaporized and escape through the leak hole leaving a stream of bubbles in the water.

The samples of welded titanium capsules were took under leak test and no bubbles detected in the water due to there were any liquid nitrogen inside the seed.

4. RESULTS

4.1. Core production

It was observed that the adsorption on single coated silver wire was consistent and quantitative, while the adsorption in bare silver wires which coated with palladium in optimal composition was poor. The percentage of ^{125}I adsorption with 10 µl KI .03 M at 60^0C was about 83% compare to about 65 %when we used 20 µl KI .03 M as a carrier at 80 ^0C. It was found that the percentage of adsorbed ^{125}I increased by increasing amount of carrier in range of 1-30 µg in volume of 10 µl. The result of autoradiography showed that the distribution of activity on the palladium coated silver wire was uniform. Main purpose of this experiment was to prepare sources with activity of about 148 MBq.

4.2. Uniformity of activity and dose distribution

In this experiment, error of obtained dose is 20%, and wavelength is 660 nm. The results indicate that nonuniformity of activity around the core is about 8%. If we use the point source approximation, dose rate at distance of 1cm for active core is 1.133 cGy/hr/mCi. In table II the result of film dosimetry is shown, Y axis is along of core length.

5. CONCLUSION

5.1. Core production and welding

The results showed that palladous chloride treated wire were more suitable for adsorption of ^{125}I than plain silver wires. Unlike simple physical adsorption of radio iodide on plain silver wires, under the same experimental conditions, the nature of adsorption of Pd treated wires seemed to follow chemiadsorption pattern which showed linear relation between amount of initial used radio iodine and adsorbed ^{125}I in range of micromoles. The stability of ^{125}I on Pd coated silver wire may be due to the formation of insoluble Palladous Iodide on the surface of the wire in water and thus may account for low leachability. It was observed that the volume and concentration of carrier, time and temperature were critical factors for adsorption. Test results of welded samples confirmed the suitable selection of the welding parameters for laser welding, and the sources can be used after careful quality control of ^{125}I sources. Experiments shows that higher activity can be achievable which needs supplementary works in this area.

5.2. Dosimetry of ^{125}I Core using MD-55 GAFCHEROMIC film

The results of film dosimetery indicate that this method can be used for determining nonuniformity of activity of the core as a routine method.

Part B: ^{192}Ir PRODUCTION

6. PRODUCTION

For construction of intracavitary source we use a cylindrical pellet of ^{192}Ir with 3 mm diameter and 0.3 mm length and maximum activity about 10 Ci for HDR machine sample which design and constructed for one of the hospitals in Iran (Fig.13). We welded stainless steel capsule with 4.6 mm outer diameter and 6.5 mm length, to 164 cm flexible cable (Fig.14). We test this joint by tensile test in several times. Its resistance against tensile was over standard level (over 1000 N). Then we sealed Iridium pellet in stainless steal capsule by argon welding. Finally we test the sources according to ISO 9978 and ISO 1677 standard, for leakage, impact, temperature vibration and puncture tests.

FIG. 13. HDR machine sample made in Iran

FIG. 14. ^{192}Ir sources produced with TIG welding for HDR machine.

7. DOSIMETRY

We used a PTW 0.6 cc farmer chamber 30001 that was calibrated in SSDL for 250 kV X ray and ^{60}Co beam. We calculated Iridium calibration factor from IAEA-TECDOC-1079: Calibration of Brachytherapy Sources.

$$N_{k,Ir} = 0.8\, N_{k,250kev} + 0.2\, N_{k,Co} \quad (1)$$

So we measured Air Kerma Rate by:

$$K(T)_{air} = (M - M_L) N_{\underline{k}} . C_{t,p} . P_{gradient} . A_{ion} . P_{ion} / T \quad (2)$$

Where:

- M: Measuring in term of charge.
- M_L: Leakage charge
- T: measuring time about 300 Sec.
- $N_{k\,:}$ Calibration factor
- $C_{t,p}$: temperature and pressure factor.

A_{ion} and P_{ion}(recombination correction factor) and $P_{gradient}$ (displacement factor) assumed be 1, beacuse their effect were considered in calibration. To minimizing attenuation and scattering we measured kerma rate in air and dose rate in water phantom at 10-20 cm from source center, in center of room. So we converted air kerma rate to 1 m from source center by inverse square law.

We developed a treatment planning software based on point source dose calculation formula TG-43, with using average anisotropy factor and meisberger polynomial for tissue correction factor. The results of calculated and measured dose rate have less than %5 differences together (Figs.15 and Fig. 16).

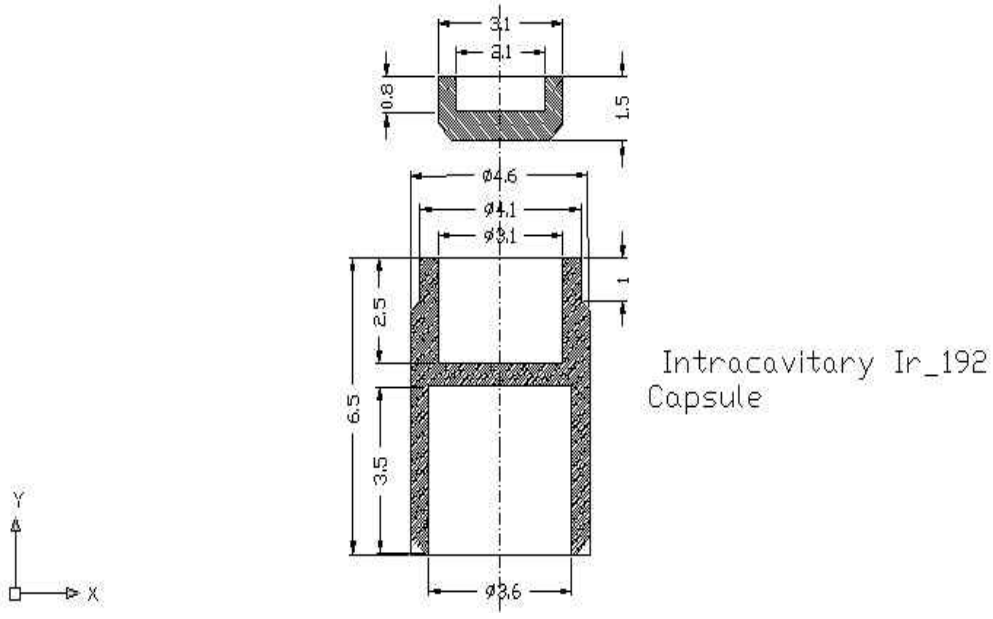

FIG. 15. Dimension of ^{192}Ir sources.

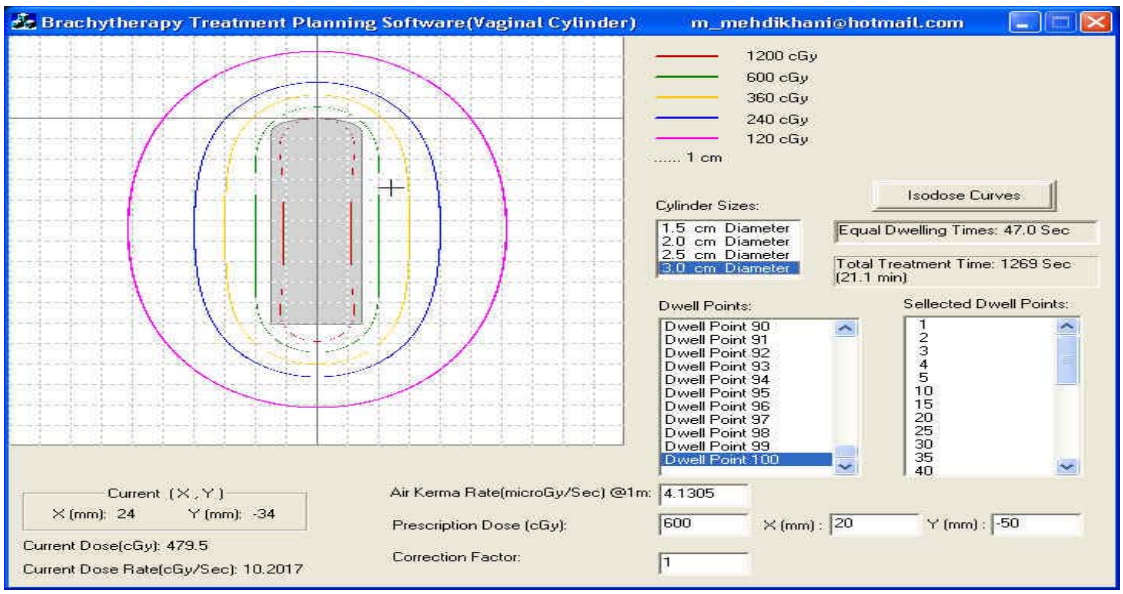

FIG. 16. Treatment planning, Isodose for ^{192}Ir HDR machine.

Finally for first radiobiological research the dose of 620 cGy was considered as lethal dose, the whole body dose of 400 cGy was applied to the mouse. Approximately 18 hr after irradiation first group of animals began to die.

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^{125}I SUBLIMATION/CHEMOSORPTION TECHNIQUE FOR MINIATURE SOURCE CORE PRODUCTION

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Abstract

The report describes methodology and results of laboratory experiments on chemosorption of molecular iodine thermally sublimated in vacuum onto silver substrates. Optimum process parameters are determined with a purpose to produce active cores of miniature ^{125}I sources for brachytherapy. The main feature of the undertaken study as compared to other works on preparation of active core for sealed iodine-125 sources was the application of vacuum process, which includes thermal sublimation of free iodine and its chemosorption on a silver substrate. The iodine layer formed by sublimation/chemosorption on silver substrate showed good time-temperature stability. No iodine escape was observed after 72 hours at 35-40^{o}C in air, and measurable loss of activity during 5 minute heating in vacuum starts from ~470^{o}C. Leachability of deposited iodine was below 0.1% in all cases. For efficient production of ^{125}I radioactive cores, the chemosorption/sublimation unit should have minimum "parasitic" surface area and maximum substrate surface area. The acceptable structural materials for the unit are fused quartz, stainless steel and pure aluminum. The results of the present study allow concluding that iodine sublimation/chemosorption technique is feasible for production of cores for miniature ^{125}I sources with high activity.

1. INTRODUCTION

Some of radioactive sources for medicine, including ^{125}I sources, are based on the active core production from the initial radioactive preparations. This require efficient fixation of the radionuclide in the core. The technique should be developed to provide sufficient activity, required activity distribution, strong binding and radionuclidic purity of the core.

World production of the miniature sealed radioactive sources for brachytherapy is concentrated in a few commercial companies. As can be found in available literature, they use different design of an active core of the seed with usual activity from about 0.2 to 40 mCi:

- Amersham's capsule contains ^{125}I "coated" or "adsorbed" onto a small silver rod;
- North American Scientific uses ion exchange resin beads with ^{125}I absorbed;
- DRAXIMAGE's capsule contains ceramic beads with ^{125}I;
- BEBIG uses porous ceramic tube loaded with silver iodide;
- Implant Sciences/MedTec uses ceramic tube containing ^{125}I buried beneath the surface;
- IBt's seed contains thin layer of ^{125}I on the outer surface of inner titanium tube.

Respectively, different technical approaches are applied for core production, from usual sorption to ion implantation. Commercial companies do not release the details of technology, but obviously the most common are various versions of sorption or chemosorption.

One of the past developments of the Institute of Nuclear Physics in Almaty was a technique for production of high-activity isotopic sources for precise nuclear spectroscopy. For preparation of high quality high active sources, a method of fractional sublimation combined with chemosorption was applied. The main advantages of this method are technical simplicity, purity of the final source, due to multiple sorption/desorption process, and a strong selective bonding of the needed radioactive element to a substrate.

The requirements to radioactive sources for precise nuclear spectroscopy and to miniature sources for therapy are very similar, as it is clear from the following comparison table (Table I).

The objective of the proposed research is to use the existing experience for development of the technique for production of an active core of miniature ^{125}I source.

Generally, sublimation/chemosorption process includes three main stages:

- Fractional sublimation from the irradiated target material;
- Transport to the substrate;
- Trapping on the substrate.

Research tasks were to develop an experimental facility for vacuum sublimation/chemosorption of iodine, to determine the features of the process and characteristics of deposited iodine layer. The main goal was to develop a method for preparation of active cores for miniature ^{125}I brachytherapy seeds (a few square millimeters surface area), and to approach to feasible batch production technology. The experiments were carried out mainly with ^{131}I-labeled natural iodine with calibrated specific activity, and further re-calculation of the results to carrier-free ^{125}I.

TABLE I. COMPARISON OF REQUIREMENTS TO LOW PHOTON ENERGY RADIOACTIVE SOURCES FOR NUCLEAR SPECTROSCOPY AND BRACHYTHERAPY.

Radioactive sources for precise nuclear spectroscopy		Miniature radioactive sources for therapy
High total activity (dozens of mCi)	=	High total activity
High specific activity	=	High specific activity
High radionuclidic purity	=	High radionuclidic purity
Uniform activity distribution on a substrate surface	≈	Controlled activity distribution in the source core
Thin active layer on a substrate		Thin active layer allows to minimize self-absorption

2. EXPERIMENT AND RESULTS

2.1. Preparation of ^{131}I-labeled free iodine

A simple technique was developed for preparation of free iodine with calibrated low specific activity of ^{131}I:

2 milliliters of water solution of KI (20 mg of natural iodine per ml) in 10-ml vial were mixed with 0.5 ml of diluted water solution of $Na^{131}I$ (with usual total activity about 1-3 MBq of carrier-free ^{131}I), 2 ml of diethyl ether and a few drops of acidic acid. The vial was closed with rubber stopper, shaken for a few minutes and "aged" at light for 1 day. Iodide ions converted to molecular iodine by chemical reaction

$$4KI + O_2 + 4CH_3COOH \rightarrow 2I_2 + 2H_2O + 4CH_3COOK,$$

and the molecular iodine was extracted by ether. Organic phase was separated from water solution, and the activity was measured in both water and organic phases. With the assumption of the proportional distribution of natural and radioactive iodine between phases, the values of mass and concentration of free iodine in the ether were calculated.

The ether with extracted labeled iodine (typically about 10 mg of iodine in 2 ml of the ether) was stored in closed vial. To prepare a source for sublimation, a portion of extract was put into small platinum cup. The ether evaporated in a few seconds, and the cap with deposited iodine was loaded into the experimental facility for sublimation.

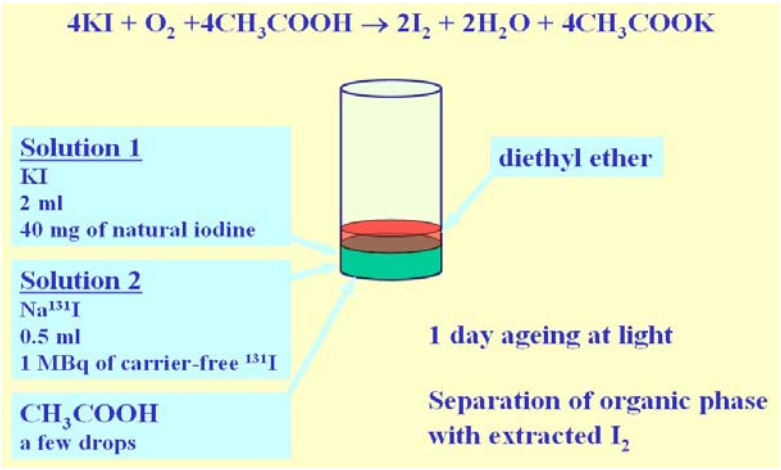
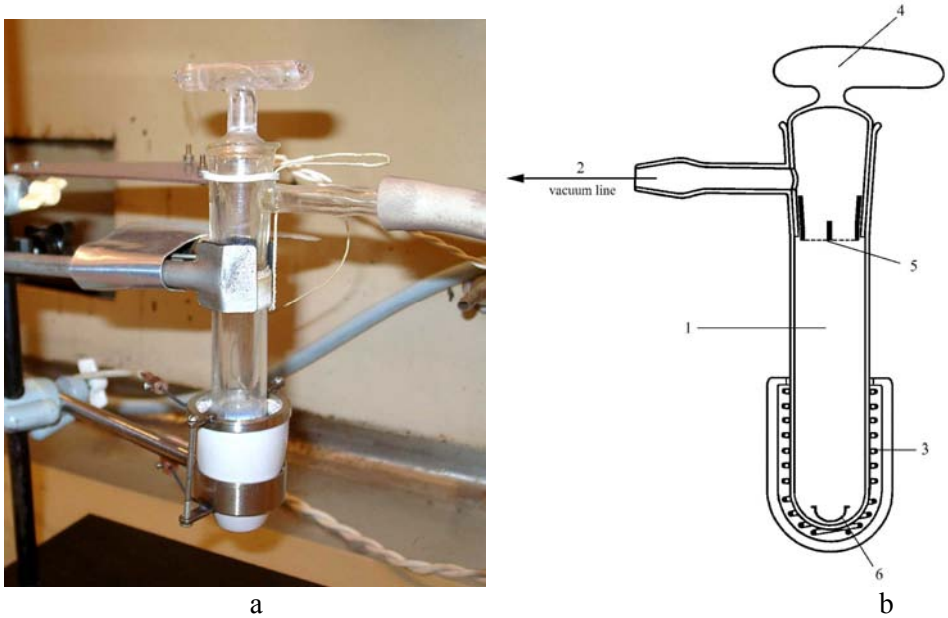


FIG. 1. Scheme of preparation of ^{131}I-labeled free Iodine.

2.2. First version of Iodine sublimation facility

For study of iodine sublimation/chemosorption, a special experimental facility (Fig.2) was designed, manufactured and mounted in the fume box equipped with additional carbon filter to trap the released iodine. The principal components of the facility are closed ampoule with evacuated work volume (1) and heater (3). The ampoule is made of fused quartz, which has low sorption ability of Astatine (chemical analog of Iodine).



a b

FIG. 2. First version of experimental facility for study of iodine sublimation/chemosorption:(a) Photo; (b) Schematic drawing.1 - quartz ampoule, 2 - vacuum line, 3 - electric heater, 4 - valve plug,5 - substrate sample, 6 - platinum cup with starting labeled iodine.

The platinum cup with labeled iodine is loaded into the ampoule through the hole on the top, when the valve plug is removed. Substrate sample is loaded into the sample holder, and the sample holder is fixed to the valve plug. Evacuation of the system by mechanical vacuum pump up to 2.7 Pa takes about 0.5 minutes.

After the required vacuum is reached, the valve is closed, and the bottom part of the quartz vessel is heated by the electric heater. The temperature in different sections of the system was measured by chromel-alumel thermocouples.

For repeatability, every experiment started from the ambient temperature (20^{o}C). The measured time dependencies of the cup temperature for different heating current are shown at Fig. 3.

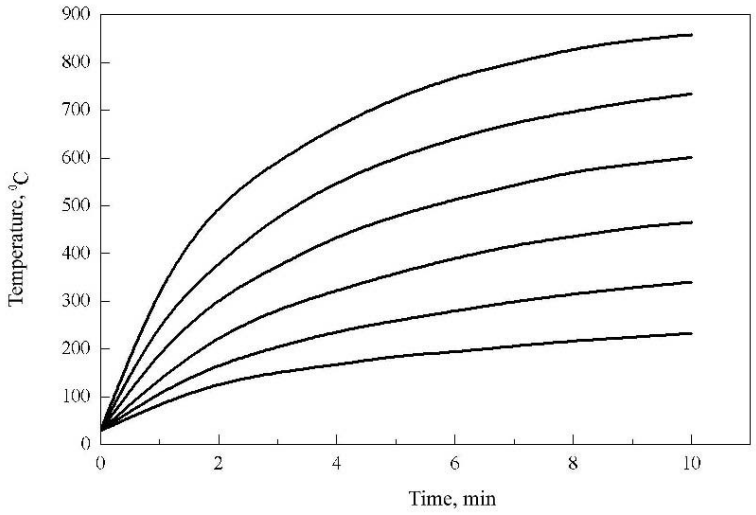


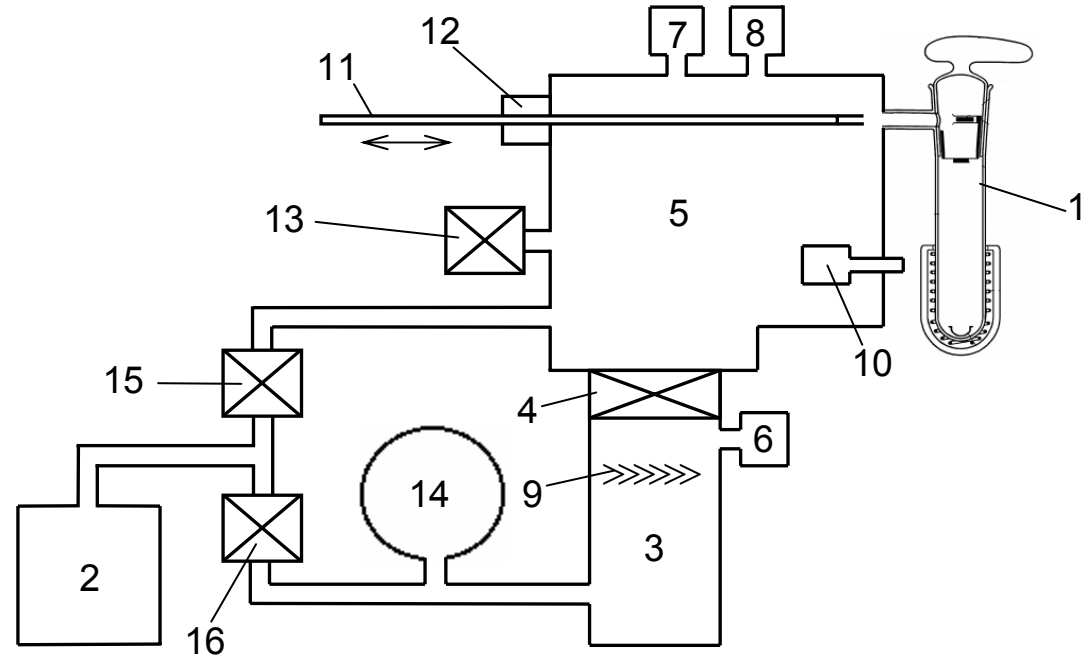
FIG. 3. Time dependence of platinum cup temperature for various values of heating current.

The values of temperature given hereinafter correspond to final (maximum) temperature in the specific experiment.

2.3. Modified high vacuum sublimation facility

In preliminary experiments with iodine sublimation/chemosorption under low vacuum conditions (10^{-1}-10^{-2} Torr), it was found that sorption parameters strongly depend on vacuum level. For this reason, the facility was modified to provide higher vacuum. In order to study sorption capacity of "fresh" silver surface deposited in vacuum, the facility was also equipped with silver evaporation chamber. Schematic drawing of the facility is shown in Fig. 4.

Relation between vacuum levels in quartz ampoule (1) and in vacuum chamber (5) was determined by using the additional ionization vacuum gage installed instead of valve plug on the quartz ampoule. Owing to application of polyphenil ether and liquid nitrogen trap (9) in the diffusion pump (3), vacuum level of $3 \cdot 10^{-7}$ Torr was reached in vacuum chamber, which corresponded to $1 \cdot 10^{-6}$ Torr in quartz ampoule. In order to obtain clean "fresh" silver surface on a substrate, the vacuum chamber is equipped with silver evaporation unit (10). Silver evaporated from the evaporation unit in vacuum is deposited onto the substrate fixed on right tip of a sample travel rod (11). After silver deposition, the travel rod moves the substrate into quartz ampoule, without loss of vacuum. Leak valve (13) is used to adjust vacuum in the system to the required level.



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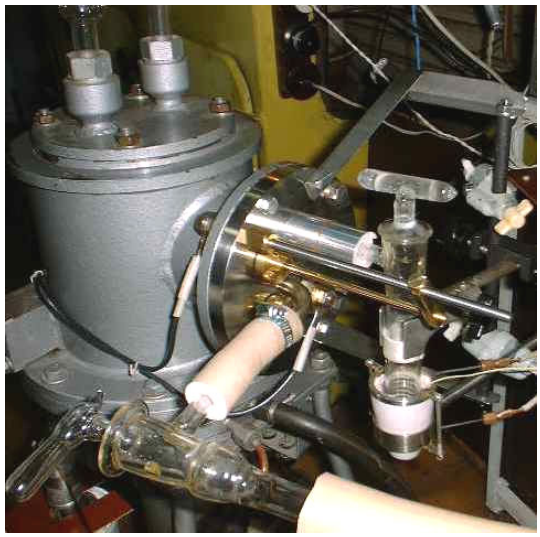

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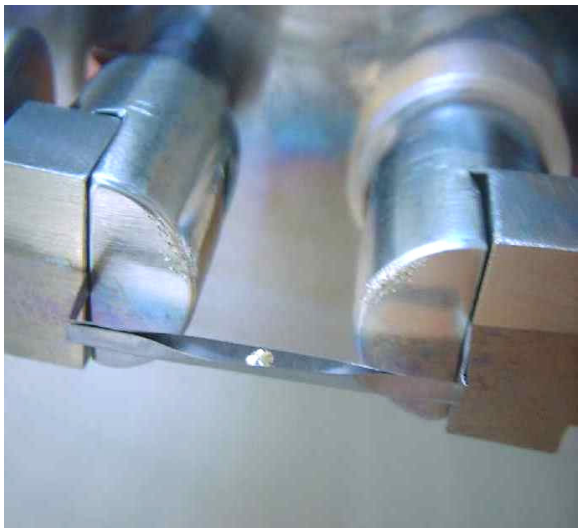


c

FIG. 4. Modified vacuum sublimation facility.

a) Schematic diagram: 1 - quartz ampoule, 2 - fore-vacuum pump, 3 - diffusion pump, 4 - gate valve, 5 - vacuum chamber, 6,7 - thermocouple vacuum gauges, 8 - ionization vacuum gauge, 9 - liquid nitrogen trap, 10 - silver evaporation unit, 11 - sample travel rod, 12 - movable vacuum seal, 13 - leak valve, 14 - rough-vacuum tank, 15,16 - vacuum valves.

b) General view of vacuum chamber.

c) Silver evaporator.

After preliminary experiments, in order to decrease substrate temperature and to allow simultaneous processing of multiple substrates the quartz ampoule was equipped with lateral pipe (sorption chamber) protected from direct heating by heat screen, a shown at Fig. 5.

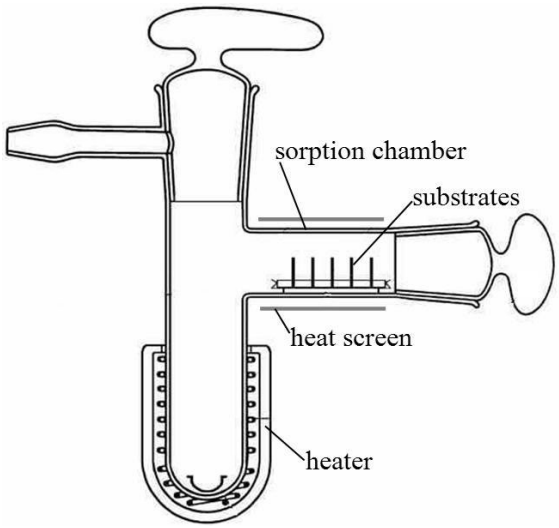
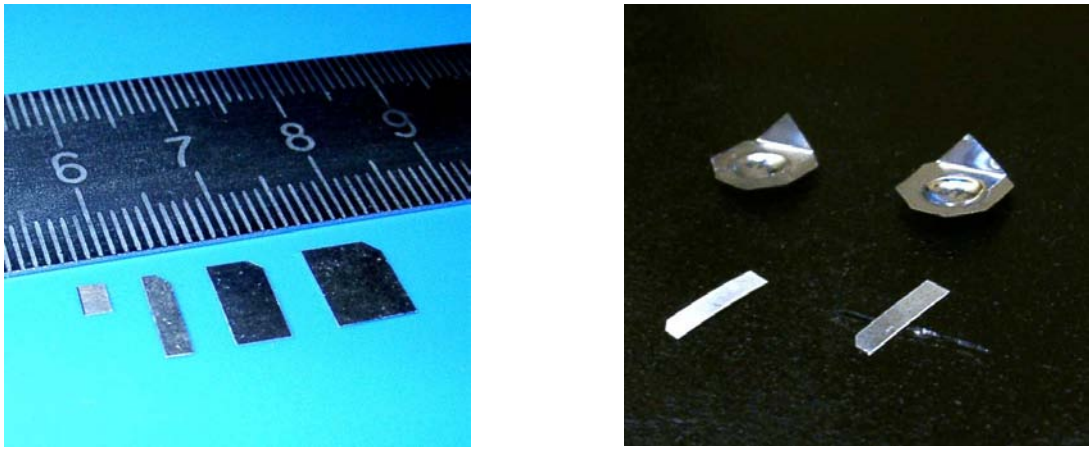


FIG. 5. Modified quartz ampoule with sorption chamber.

2.4. Shape and size of substrate samples used

Usual core of miniature therapeutic source with ^{125}I is a cylinder or sphere with surface area about 5 square millimeters. Such a small size causes difficulties and significant measurement errors in multiple experiments on chemosorption. To check the dependence of iodine chemosorption on shape and size of the substrate, preliminary experiments have been carried out with spherical, cylindrical and flat foil silver samples (12 mm^2 surface area in all of these cases), and also with foil samples of different size (Fig. 6). It was found that the effect of sample shape on saturated iodine activity is very low, a few percents, and effect of sample size is zero. Therefore, taking into account handling and measurement convenience, flat silver foils (99.9% Ag, 70 μm thick) with 12-16 mm^2 surface area were taken as a "standard" substrate sample in experimental determination of principal features of vacuum sublimation/chemosorption. In the experiments on simultaneous processing of multiple substrates, 0.6 mm diameter silver wire samples were also used.



a **b**

FIG. 6. a. Flat foil samples of different sizes used in experiments;b. Silver substrate samples and platinum cups for iodine sublimation.

2.5. Temperature dependence of Iodine sublimation from the surface of platinum cup

Surface of platinum cup is cleaned by immersion into a mixture of HNO_3 и HCl (1:3) for 3 minutes and subsequent intense washing by twice distilled water. A portion of ether containing known quantity of Iodine with known specific activity is put into the dried cup. After complete evaporation of the ether (a few seconds) the cup with iodine was loaded into the bottom of quartz ampoule.

The ampoule is evacuated by vacuum pump, and the heater is switched "on". The system is heated for 5 minutes, and a final temperature is recorded as a temperature of the specific experiment. After 5 minutes, the heater is switched "off" and removed from the ampoule. The ampoule is cooled in air for 1 minute and opened. The platinum cup is extracted from the ampoule, the remaining amount of iodine in the cup is determined by activity measurement, and a fraction of sublimated iodine is calculated. The measurements were carried out for various temperatures in the range from 20°C to 470°C in vacuum $2 \cdot 10^{-2}$ Torr. The resulted temperature dependence of sublimated iodine yield is shown in Fig. 7.

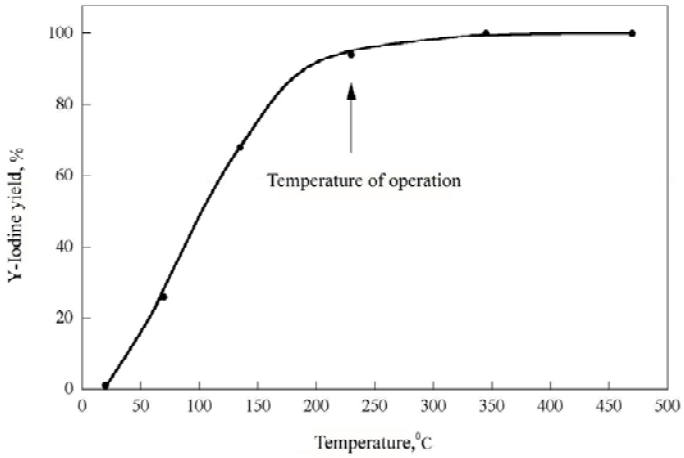


FIG. 7. Iodine sublimation yield depending on final temperature of 5-minute heating.

Also, the effect of gas pressure in the system on sublimation yield was checked, and it was found that deviation from the dependence shown in Fig. 7 did not exceed measurement error, ~ 2%, at $1 \cdot 10^{-1}$ Torr air pressure.

2.6. Determination of iodine chemosorption saturation limit for silver surface

In most of experiments for determination of saturation limit, silver foil samples with 12 mm^2 surface area were used as substrates. The samples were cleaned by immersion for 1 minute into fresh mixture of NH_3 and H_2O (1:1) with some H_2O_2 added, and subsequent washing in twice-distilled water. The dried sample was loaded into sample holder fixed on the plug of quartz ampoule, the plug closed the ampoule with iodine in platinum cup on the bottom, and the system was evacuated up to required vacuum (it took about 1 minute). The bottom section of the ampoule was heated for 5 minutes up to 230°C then the ampoule was cooled in air and opened.

Activity of ^{131}I on the silver substrate was measured then the sublimation cycle was repeated with the same substrate and fresh portion of labeled iodine in the platinum cup, up to saturation of the substrate. Accumulated activity of ^{131}I on the substrate was converted into the mass of carrier iodine and finally into the activity of carrier-free ^{125}I. Then, the experiment was repeated with larger initial amount of iodine in platinum cup which was knowingly exceeding the amount required for saturation. The saturated activities for multi-step and single-step procedures were found to be the same.

Study of time dependence of iodine sorption on silver substrate at fixed temperature gave an upper estimate of time needed to reach the equilibrium state in work volume of the ampoule. This time

proved to be not more than 20 seconds. So, 5 minute heating time was quite enough to reach equilibrium.

2.6.1. Effect of substrate temperature on iodine chemosorption saturation limit

As it was proposed that chemosorption saturation limit should depend on substrate temperature, the saturation was measured at three different temperatures, provided by location of the sample holder at different distances from the heater. At the level of lower edge of the plug, the temperature was 70°C, at the upper edge of the heater it was 230°C, and in the middle between plug and upper edge of heater it was 130°C. The results are presented in the Table II.

TABLE II. EFFECT OF SILVER SUBSTRATE TEMPERATURE ON SATURATED ACTIVITY.

Silver substrate temperature, °C	Saturated surface activity density, converted to mCi/mm^2 of ^{125}I	
	vacuum $2\cdot10^{-2}$ Torr	vacuum $1\cdot10^{-4}$ Torr
70	12.4 ± 0.4	16.5 ± 0.6
130	11.9 ± 0.4	15.5 ± 0.6
230	9.2 ± 0.4	12.5 ± 0.6

As one can see from table, the temperature dependence of saturated activity is similar for various vacuum conditions: increase of substrate temperature results in decrease of saturated activity.

2.6.2. Effect of system vacuum on iodine chemosorption saturation limit

Saturated activity of the substrate strongly depends on residual gas pressure (vacuum level) in working volume. This dependence was checked experimentally, and the results are presented in the Fig. 8 (mean value of 5 measurements for each vacuum level, at substrate temperature 70°C).

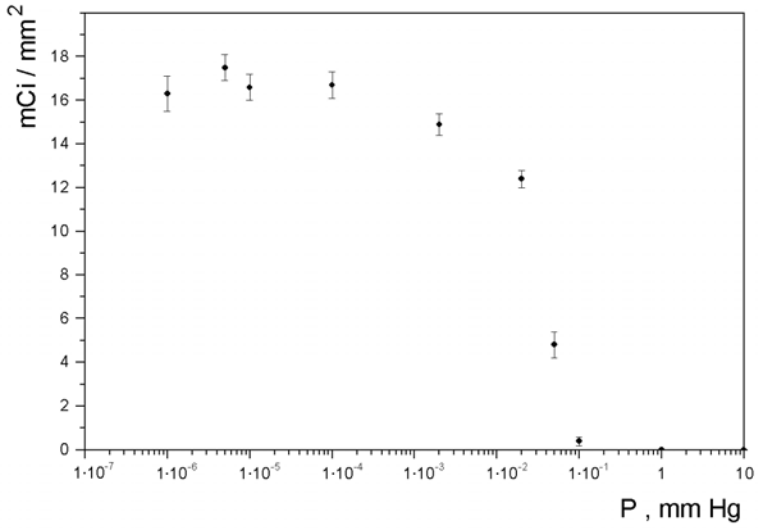


FIG. 8. Saturated density of ^{125}I activity versus residual air pressure in sublimation ampoule.

As one can see from the graph, the efficiency of iodine sorption grows rapidly with pressure decrease at low vacuum region, and remains nearly constant (16-17 mCi/mm^2) with pressure decrease below $1\cdot10^{-4}$ Torr.

Saturated specific activity at high vacuum is about 40% better than in low vacuum, so high vacuum conditions are preferable. At the same time there is no sense to go to vacuum better than $1\cdot10^{4}$ Torr, which is easily achievable with any diffusion pump.

2.6.3. Effect of substrate surface condition on iodine chemosorption saturation limit

Another parameter affecting the process of chemosorption should be a condition of the substrate surface, which depends on method of its preparation. For this reason, three different types of silver substrates were used in the experiments:

- the samples prepared by mechanical rolling with subsequent chemical cleaning,
- the samples prepared by vacuum vapor deposition without exposure in air,
- the samples prepared by vacuum vapor deposition after 25-30 minute exposure in air.

The results are presented in Table III.

TABLE III. EFFECT OF SUBSTRATE PREPARATION METHOD ON SATURATED SURFACE ACTIVITY.

Sample preparation method	Saturated surface activity density, converted to mCi of ^{125}I per mm^2
Mechanical rolling + chemical cleaning	16.5 ± 0.6
Evaporation in high vacuum ("fresh" surface)	23.5 ± 0.7
Evaporation in high vacuum + 30 min exposition in air	16.5 ± 0.6

No difference in saturated activity was found for mechanically rolled and vacuum-deposited samples after exposure in atmosphere. For both types of samples, some decrease of saturated activity (exceeding measurement error, ~3%) was observed when surface "aging" time was more than 1 day in air at ambient temperature. "Fresh" vacuum-deposited surface showed the highest sorption capacity, which was about 40% higher than that for the samples exposed in air.

2.7. Determination of sorbed portion of sublimated iodine

Beside the saturated activity density of ^{125}I on the surface, another important characteristic is an efficiency of use of sublimated iodine. To determine this efficiency, the experiments were performed on determination of portion of sublimated iodine trapped by the substrate surface. In these experiments, the quantity of "initial" labeled iodine in platinum cup was taken small enough to avoid the substrate saturation. Final activity of ^{131}I tracer on the substrate was measured and compared to the initial activity of sublimated iodine.

Three sets of measurements were done, and the average portion of iodine chemosorbed on silver substrate was found to be (24 ± 2)%. It should be noted that surface area of silver was about 3 orders of magnitude times smaller than the internal surface of quartz ampoule.

Similar experiment was carried out with heating of a smaller part of quartz ampoule, only directly near the platinum cup. The portion of iodine trapped on a silver substrate is (11 ± 1)% in this case.

The same study was repeated for substrate made of copper (99.9% Cu), and the results were (18 ± 2)% and (7 ± 1)%, respectively.

A possibility of enhancement of use of sublimated iodine by heating of system walls was also checked. As expected, this gives a positive result. Almost complete desorption of iodine from quartz surface (and its redistribution to silver substrate) was observed at ~500^{o}C.

2.8. Temperature-time stability of iodine layer on substrate

Application of iodine-125 sources in therapy requires temperature and time stability of iodine layer in the core, to provide a stable radiation dose rate distribution around the source. Temperature stability is important also because of welding used for sealing of source capsule.

To study temperature stability, the iodine release from silver substrate was measured at various temperatures. In these experiments, platinum cup with iodine on the bottom of quartz ampoule was replaced by 12 mm^2 silver substrate saturated by iodine with known activity of ^{131}I tracer. "Fresh" clean silver foil was located in upper part of the ampoule, as potential trap for released iodine. The substrate was heated for 5 minutes in vacuum $2 \cdot 10^{-2}$ Torr up to the required temperature. After cooling, the remaining activity of substrate was measured and escaped portion of the activity was calculated. The results are shown in Fig. 9(a).

A separate experiment was conducted to check time stability of saturated iodine layer on silver substrate at 35-40^{o}C in atmosphere. The measurements performed after 24, 48 and 72 hours showed no iodine escape (taking into account a normal radioactive decay of ^{131}I tracer) with accuracy ~1%.

Similar experiments were carried out also with copper substrate samples (99.9% Cu) reasoning from the similarity of chemical behavior of copper and silver. 12 mm^2 copper samples were saturated by iodine in the same way as silver substrates, and then iodine release in vacuum was studied at various temperatures. The results are presented in Fig. 9(b). Measurements of iodine escape at 35-40^{o}C in air after 24, 48 and 72 hours, as for silver, gave zero results.

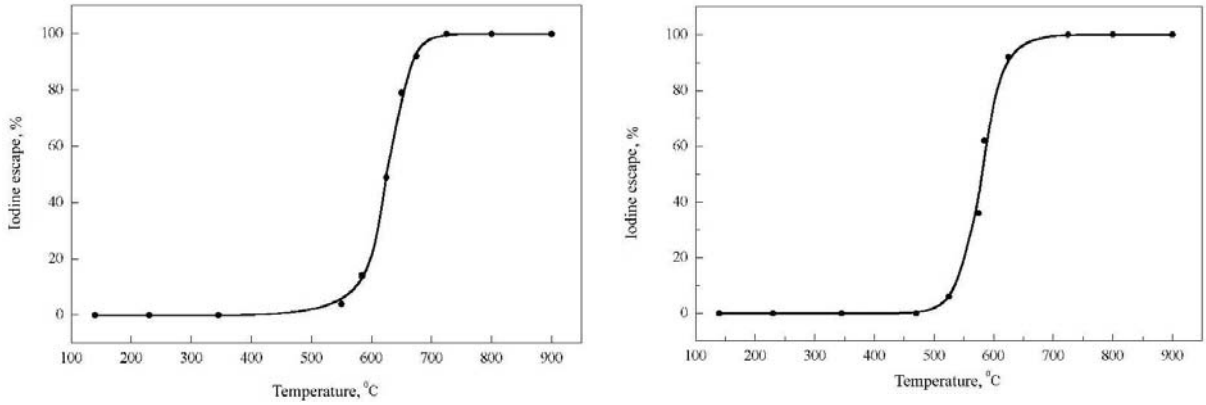


FIG. 9. Temperature dependence of iodine escape in vacuum (5-minutes heating): (a) - from silver substrate; (b) - from copper substrate.

2.9. Leachability of deposited iodine from substrate

Leachability of deposited iodine layer was checked by immersion the samples into bi-distilled water at 20°C temperature for 24 hours. In all cases, the leached activity was found to be less than 0.1% for silver substrates. For comparison, about 20% of deposited iodine activity leaked to water in the same conditions from copper substrates.

2.10. Uniformity of the deposited iodine layer

Uniformity of deposition was studied as difference of iodine activity at multiple substrate samples in one batch, and also as iodine activity distribution along the single wire sample.

2.10.1. Multiple samples.

The measurements were done at flat foil silver samples with dimensions 4 mm x 4 mm assembled in array, at vacuum $1 \cdot 10^{-4}$ Torr and deposited activity about 10% of saturated activity. It was found that difference of activity deposited on different foils is below 3% until the distance between the foils is larger than 1.3 mm, i.e. about 1/3 of sample size.

2.10.2. Distribution along wire sample.

Experiments were carried out using 6 mm long silver samples with diameter 0.6 mm installed at 0.6 mm distance between the samples. Deposited activity of ^{125}I was about 10% of saturation limit. Activity distribution along single wire piece was determined by measurement of conversion electrons using beta-spectrometer with 1 mm collimator slit. Position of slit was moved by micrometer screw. The activity was found to be uniform along the wire piece with 3% error (see Fig. 10).

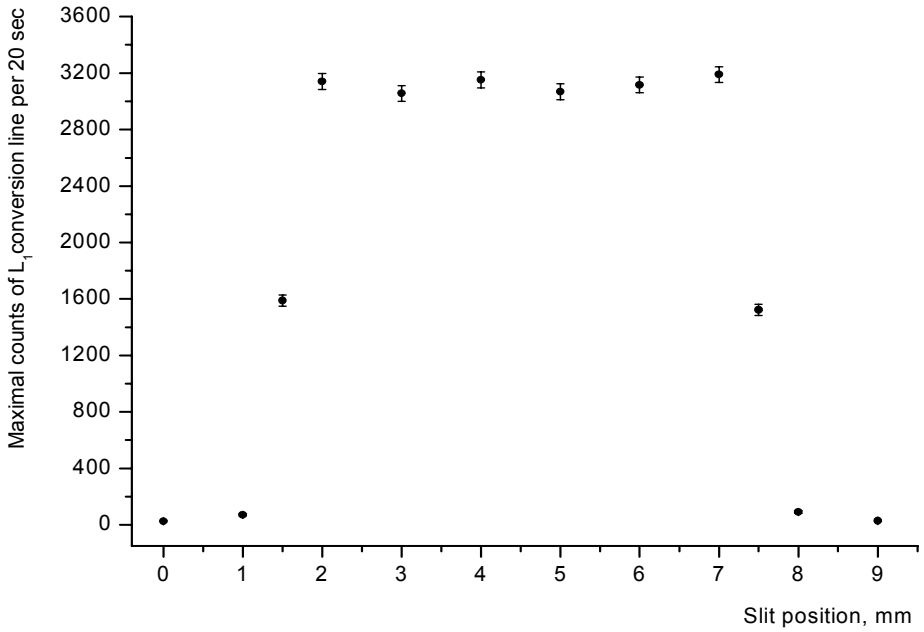


FIG. 10. Iodine-125 activity distribution along 6 mm long silver wire measured by conversion electron scan with 1 mm collimator slit.

2.11. Selection of materials for sublimation facility

It is clear that the best structural materials for construction of iodine sublimation-chemosorption unit are those with minimum sorption capacity. Several commonly used materials were checked for sorption of iodine in vacuum $1 \cdot 10^{-4}$ Torr, to study their suitability for this purpose. The amount of sublimated iodine in each experiment was exceeding the amount needed for saturation of the substrate. Saturated surface activity density was compared with the value for fused quartz. The results are presented in Table IV.

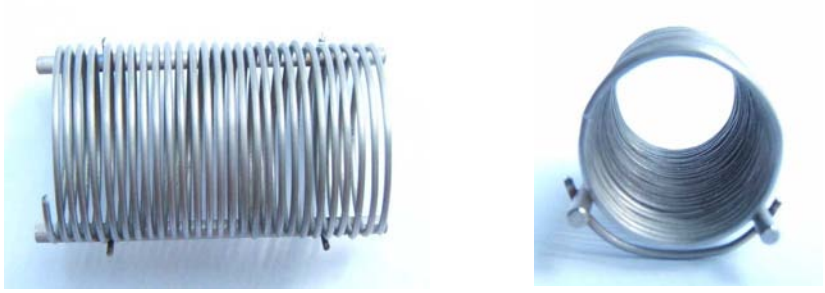
TABLE IV. RELATIVE IODINE SORPTION CAPACITY OF SEVERAL STRUCTURAL MATERIALS

Material	Relative sorption capacity, %
Silver	100
Fused quartz	0.8
Non-fused quartz	6
Molybdenic glass	6
Stainless steel	5
Pure aluminum	4
Duraluminum	7
Al-Mg-Si alloy	9
Fluoroplastics, various grades	21-27
Alundum ceramics	29

As it is seen from the table, the lowest sorption capacity was found for fused quartz. Non-fused quartz showed significantly higher value, as well as other tested materials. A limited amount of stainless steel and pure aluminum components may be used in the sublimation unit. Fluoroplastics, ordinarily used in vacuum facilities, should be avoided in this specific case.

2.12. Approach to batch production of iodine-125 source cores

Batch production of iodine-125 cores for miniature brachytherapy sources requires simultaneous processing of large amount of silver substrates. One of the most effective and simplest ways to do this is usage a long silver wire in a form of coil as a substrate for iodine deposition. In the applied experimental facility, a small sorption chamber accommodated 1.5 m of 0.5 mm diameter silver wire in the form of 16 mm coil with 0.5 mm pitch and 30 mm length. This size wire after iodine deposition may be cut onto 500 pieces, each 3 mm long, which can be used as cores for the sources. The coil holder made of stainless steel provides the necessary distance of substrate from the chamber walls (see Fig. 11).



a **b**

FIG. 11. Compact wire coil for use as a substrate for iodine deposition in vacuum:a) top view, b) front view.

3. CONCLUSION

The main feature of the present study as compared to other works on preparation of active core for sealed iodine-125 sources is application of vacuum process, which includes thermal sublimation of free iodine and its chemosorption on a silver substrate.

In the usual method of production of iodine-125 by irradiation of Xenon gas target it is easy to form free iodine (ready to sublimate) precipitated on ampoule walls. So, the irradiated ampoule may be directly used as a sublimation source. For 5 minutes, a loss of free iodine from the ampoule at ambient temperature is about 1%, whereas 95% of iodine will sublimate at 230^{o}C and 100% at 400^{o}C. 230^{o}C was chosen as an operation temperature in the experiments, allowing to provide almost complete sublimation of iodine and to avoid system overheating.

In most of model experiments, the ^{131}I labeled free iodine was used, because of easier activity measurement.

Silver was taken as a substrate material for chemosorption of sublimated iodine, because it forms a strong chemical bond with iodine and is frequently used as core material for therapeutic iodine sources. Behavior of copper as a substrate material was found to be very similar to silver, however most of measured parameters were slightly worse for iodine layer on copper.

The parameter limiting activity of the radioactive source in case of chemosorption is saturated surface activity density on the core. It was found that saturation limit is about 16.5 mCi/mm^2 (610 MBq/mm^2) at substrate temperature 70^{o}C and vacuum $1 \cdot 10^{-4}$ Torr for chemically cleaned silver surface, and about 23.5 mCi/mm^2 (870 MBq/mm^2) for "fresh" vacuum-evaporated silver layer. This level of surface activity allows concentrating on a silver core of standard size brachytherapy source (0.5 mm diameter and 3 mm length) much higher ^{125}I activity than it is usually reported. A standard source will contain about 80 mCi (2.96 GBq) of ^{125}I homogeneously distributed in a very thin layer on a surface of the silver core. The uniformity of iodine distribution on a substrate is confirmed experimentally. As to kinetics of the process, it was found that 20 seconds is enough time to reach equilibrium state in the system.

The iodine layer formed by sublimation/chemosorption on silver substrate showed good time-temperature stability. No iodine escape was observed after 72 hours at 35-40^{o}C in air, and measurable loss of activity during 5-minute heating in vacuum starts from ~470^{o}C. Leachability of deposited iodine was below 0.1% in all cases.

For efficient production of ^{125}I radioactive cores, the chemosorption/sublimation unit should have minimum "parasitic" surface area and maximum substrate surface area. The acceptable structural materials for the unit are fused quartz, stainless steel and pure aluminum.

The results of the present study allow concluding that iodine sublimation/chemosorption technique is feasible for production of cores for miniature ^{125}I sources with high activity. General requirements to the process and facility can be formulated as follows:

- heating of starting iodine source above 200^{o}C;
- temperature of silver substrate below 100^{o}C;
- vacuum $1 \cdot 10^{-4}$ Torr;
- minimized internal surface area of work volume;
- possibility of warming of work volume walls;
- preferable structural materials are fused quartz, pure aluminum and stainless steel;
- a sorption chamber should accommodate a compacted long silver wire (in a form of coil or assembly of straight pieces), for batch production of the source cores.

DEVELOPMENT OF MINIATURE RADIATION SOURCES FOR MEDICAL AND NON-DESTRUCTIVE TESTING APPLICATIONS

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Abstract

This work aims to develop ^{192}Ir sources for the high dose rate brachytherapy, ^{125}I seeds for the therapeutic purpose, and ^{169}Yb sealed sources for industrial applications. ^{192}Ir sources developed at KAERI have approximately 10 Ci (370 GBq) activities. For a safety assurance, the welded joints and capsule of the sources are thoroughly tested and inspected based on the ISO2919:1999 standard. To develop the ^{125}I seed the carrying media consisting of Al_2O_3 and silver nano powder was developed as a precision assembly welding system. Two welding methods for sealing iodine-absorbed media have been developed. As a low gamma ray-emitting isotope, ^{169}Yb sources have been developed and tested for their performance. Optimal compacting and sintering conditions for the fabrication of Yb_2O_3 pellet (1mm length and 1mm diameter) are determined experimentally. Source holders for ^{169}Yb are designed and fabricated.

1. INTRODUCTION

Radioisotopes have been employed in various industrial, medical, environmental, and advanced scientific fields. Commercial applications are extended continuously with the development of the industries. Radioactive isotopes are currently used in more than 2,000 organizations in Korea, and the domestic demand is expected to grow steadily. The Korea Atomic Energy Research Institute (KAERI), which operates a research reactor (HANARO) is the sole production organization of radioisotopes (RI) in Korea. Currently KAERI has interest in the development of radioisotopes for medical applications because the medical demand is growing fast, and the supplies heavily rely on imports. Also, KAERI has interest in the development of sources of low gamma ray-emitting isotopes for industrial applications [1,2].

On behalf of these efforts, ^{192}Ir high dose rate brachytherapy sources, ^{125}I therapeutic sources, and ^{169}Yb sources have been developed or are under the development. The development of ^{192}Ir high dose rate brachytherapy sources has been accomplished. The ^{192}Ir source has the dimension of 1.1mm in diameter and approximately a 10 Ci activities. An automatic laser welding and assembling system are developed for the fabrication of the sources. For a quality assurance, welded joints of the sources are thoroughly tested and inspected. These sources are also tested for radiological, mechanical, and medical characteristics at a domestic hospital by using a remote after loading system. [3,4,5]

^{125}I seeds are currently under development. These activities include designing and manufacturing titanium caps and tubes, which are outer sealing elements of the seeds. A micro-positioning system has also been developed to assemble and to weld the sealing. Various media are considered as ^{125}I carriers, such as silver rods, ceramic rods, and ceramic/silver composites.

^{169}Yb as a low gamma ray-emitting isotope has been employed to inspect tubes and plates with relatively small dimensions. Optimal compacting conditions for the fabrication of a small size pellet (1mm length and 1mm diameter) of Yb_2O_3 powder are determined experimentally. The sources are double-sealed by an aluminium inner capsule and titanium outer capsule. To obtain the optimal welding conditions for the capsule joints, the welding characteristics have been analysed from various aspects.

The objective of the research work was the design and fabrication of seed sources, characterization of them and the development of the production technologies. In addition, automation systems for a stable production of such sources were investigated.

2. ^{192}Ir SOURCE FOR A HIGH DOSE RATE BRACHYTHERAPHY

Two types of high dose rate ^{192}Ir sources (1.1 mm and 4 mm in diameter) have been developed for medical applications. A laser welding system consisting of a laser welder and an automatic welding device was developed to fabricate the sources. Four mm sources at 10 Ci (370 GBq) activities are supplied to domestic hospitals to test the source as a replacement to the ^{60}Co brachytherapy sources. In the tests, these sources have been successfully applied for patient treatments. The technology for the production of the 4mm ^{192}Ir sources was transferred to a private company for a commercial manufacturing.

Radioactive sources for an intracavitary irradiation of a remote-after-loading are ^{60}Co, ^{137}Cs, and ^{192}Ir. ^{192}Ir source has received a great deal of attention recently for this purpose because it has advantages of relatively smaller size and lower emission energy. Since this source has a 380 keV energy level and 1.1 ~ 1.6 mm in diameter, it can be used for the treatment of blood vessels or bile ducts that have a narrow space. Hence, miniaturized ^{192}Ir sources have also been developed for remote after loading systems. A ^{192}Ir pellet (D x L = 0.6 mm x 3.5 mm, 22.6 mg, 99.9% pure) is singly encapsulated in a cylindrical stainless steel capsule (SUS 316, D x L = 1.1 mm x 6.0 mm) as shown in Fig. 1.

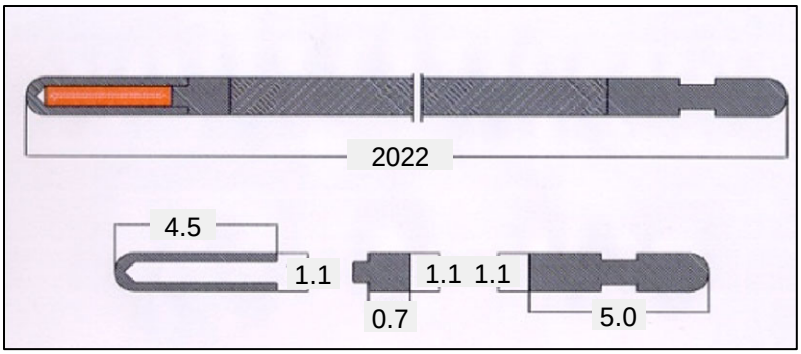


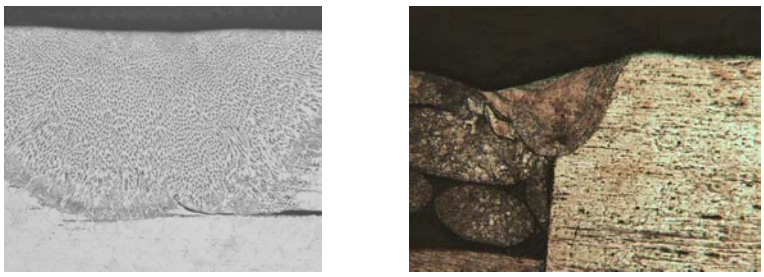
FIG. 1. Schematic drawings of ^{192}Ir brachytherapy source assembly

2.1. Technical specifications and fabrication system

One side of this capsule is welded to a flexible metal cable (D x L = 1.1 mm x 1500 mm), which has an engraved metal tail, to be installed into the loading system. Also, the cable tail has an engraved serial number and marked with a colour for type recognition. The cable used in this study withstands a permanent deformation at a 30 mm radius of gyration and a 15mm radius gyration at a 150 mm distance from the capsule tip. A mechanical test by pulling it with a 15N force for up to three minutes, all the welded parts are microscopically checked as shown in Fig. 2. Based on these test results, optimal welding conditions are determined as shown in Table I. Leak tests are also performed to inspect the 5 nCi for a removable contamination prior to shipping.

TABLE I. OPTIMAL WELDING CONDITIONS OF ND-YAG PULSE LASER FOR MANUFACTURING ^{192}Ir BRACHYTHERAPY SOURCE

Shielding gas flow rate	Input voltage	Pulse width	Frequency
High purity helium 3 ~ 4 liter/min	280 V	10 msec	8 Hz



(a) *(b)*

FIG. 2. Microstructure of welded joints; (a) joint between cap and capsule, (b) joint between source wire and cap end

Since the ^{192}Ir miniature source for brachytherapy has a relatively small size and a high specific activity, handling of the source assembly is not easy. Hence, it is important to employ an automation system to perform a precise welding and handling of the sources. The developed automatic fabrication system consists of a laser-welding machine, a cutting and welding devices for the source cable, a source assembling and welding device, a control unit, and an imaging process unit as the schematic drawings in Fig. 3 and Fig. 4 show.

2.2. Quality controls and pre-clinical study

The developed sources are tested according to ISO 2919 requirements. The tests are vacuum bubble tests after exposing the source to a low and high temperature, external pressure, and impact (Table II). The sample in thermal test is performed first in a low temperature condition (-40℃, 20 minutes) and then in a high temperature condition (600℃, 1 hr) followed by a thermal shock by a sudden quenching in water at 20 ℃. External pressure tests are performed by depressurizing it to 20 kPa and then raising the pressure to 2MPa (≒20atm) for five minutes. The leaking of the sealed capsule is determined by the weight change of the capsule due to the water intrusion during the test. Impact tests are conducted by dropping a 50g weight from a 1.0m height to the weakest point of the source capsule. No longitudinal deformation or 20-25% latitudinal expansion of the capsule is observed. The soundness of the sealing of the capsule samples from these tests are measured by a bubble test. In this bubble test, the specimens are cooled down in liquid nitrogen and added to methanol to observe the evolution of the bubbles. No bubble evolution is observed from this test. Hence, it can be concluded that the Iridium source capsule is strong enough and well-sealed.

The feasibility study of the ^{192}Ir source for HDR brachytherapy with a Microselectron applicator (Nucletron, USA) is performed. The fabricated ^{192}Ir source assembly is installed in the applicator. Four tests are performed to validate its applicability. These tests are, 1) 800 times loading/unloading test to verify the structural endurance; 2) exposure test to observe the accuracy of the source positioning; 3) tests for the source movement and the positioning accuracy of various brachytherapy applicators; and 4) evaluation of a source movement in a curved pathway. From the results of these tests, the welded joints of the source assembly do not have any crack or damage by the repeated loading/unloading cycles. The source positioning is within the allowance of 1 mm. Source assembly is able to move without any problems inside various applicators. The source does not move when the curvature is smaller than 28.95 mm of the gyration radius. Also, radiation safety equipment works normally after an installation of the developed source to the Microselectron.

Activity and anisotropy of the source assembly are measured. To measure the activity, a fabricated phantom, which has a Farmer type chamber (PTW 30006, PTW, Germany) and an endobronchial catheter is used. The endobronchial catheter makes a circle with a 10 cm radius from the centre of the PTW 30006 chamber and the source is moved inside the catheter.

The source is positioned for total dwell times, of 72 s and 12 s at 12 points with an equal interval inside the catheter, and then the charge (in nC) integrated by the chamber from the stationary source is obtained. Finally this charge is converted to an apparent activity of the ^{192}Ir source. Anisotropy for the ^{192}Ir source is obtained through dose measurements in a water-equivalent phantom. With lithium fluoride thermoluminescent detectors (1x1x6 mm^3) positioned in a polystyrene phantom, the doses at angle positions with 10°intervals from 0° to 170° are measured at distances of 5cm and 7 cm, respectively. Anisotropy for the source assembly is similar to that obtained with the Nucletron source.

TABLE II. ISO2919 CLASSIFICATION STANDARD

Test	Class	Method
Temperature	5	-40°C(20min), +600°C(1hr) and thermal shock to 20°C
External pressure	3	25 kPa absolute to 7 Mpa absolute
Impact	2	50 g from 1 m or equivalent imparted energy
Vibration	1	No test
Puncture	1	No test

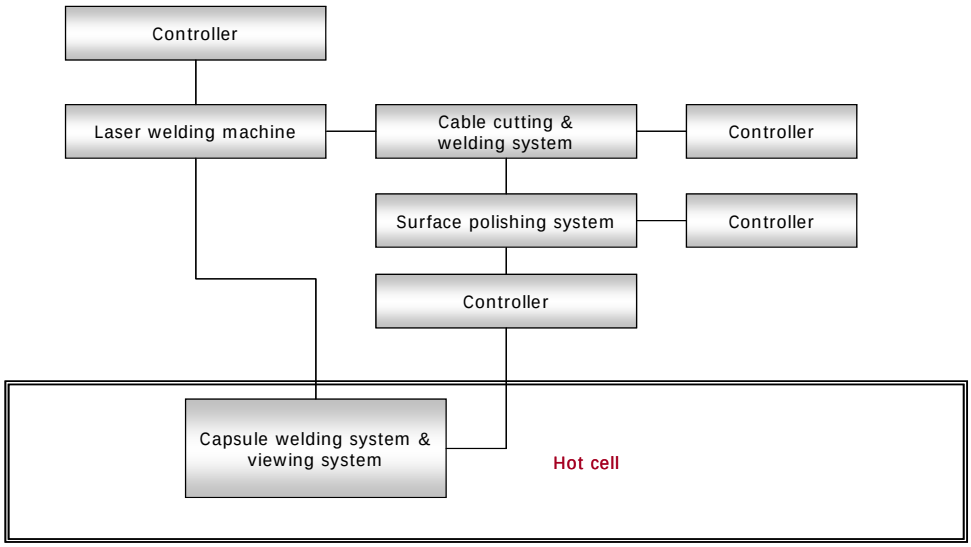
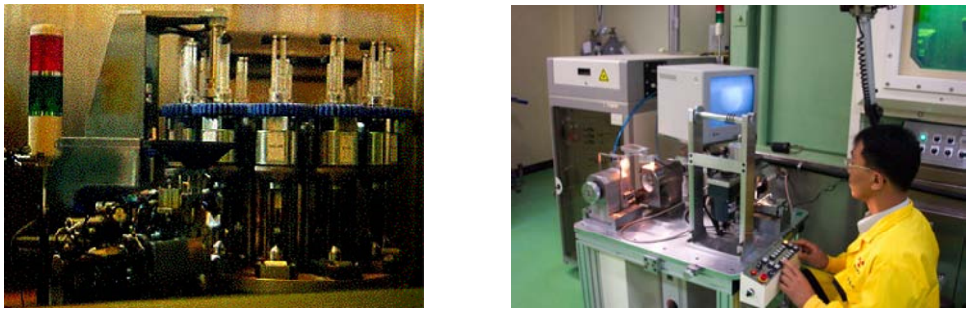


FIG. 3. Schematic diagram of the automatic fabrication system for the Ir-192 miniature source



(a) (b)

FIG. 4. Photography of the system; (a) controller, laser welding machine, and cable cutting and welding system, (b) automatic fabrication capsule welding system installed in hot cell

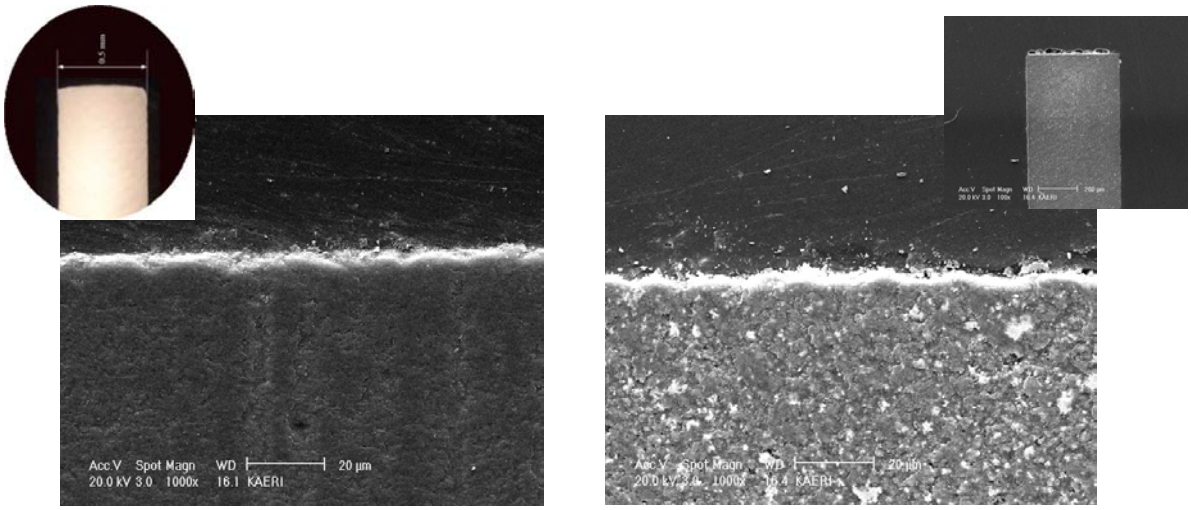
3. PRODUCTION TECHNOLOGY OF THE ^{125}I SEED

During the last decade, brachytherapy has emerged as a novel treatment method for localized eye and prostate cancer. Brachytherapy employs radionuclides that emit principally low-energy gamma rays. It occupies a recognized place in the radiation therapy modality. The radionuclide ^{125}I is primarily chosen for this purpose.

Radioactive ^{125}I can be produced by bombarding neutrons on ^{124}Xe. ^{125}I decays via an electron capture to the first excited state of ^{125}Te with a half-life of 59.4 days. Subsequently, the decay proceeds principally by an internal conversion resulting in the emission of 27 keV and 31 keV X rays and 35 keV γ -ray emissions. ^{125}I sources for the purpose of brachytherapy are fabricated in the form of seeds, 4.5 mm long and 0.8 mm in diameter.

3.1. Development the adsorption technique

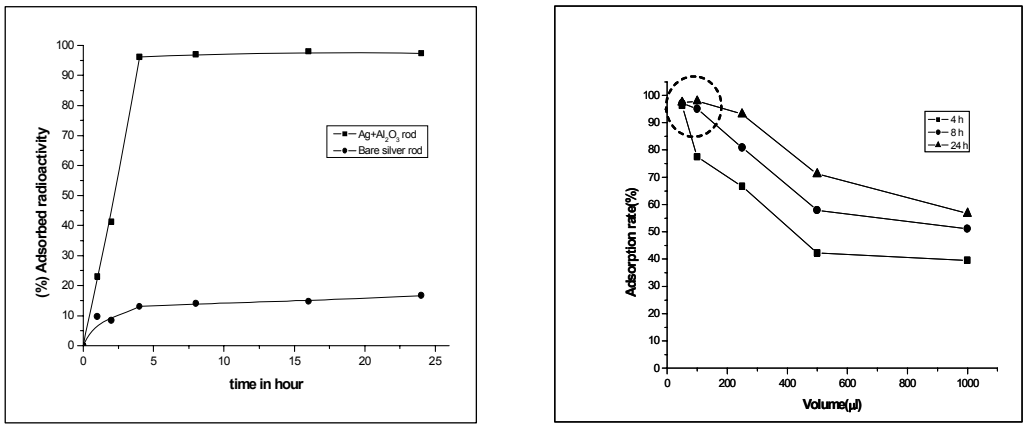
For the ^{125}I brachytherapy seed, a carrying media consisting of Al_2O_3 and silver nano powder is developed. In this material, approximately 15 wt% of Ag is contained. The porosity of the material is approximately 10%. The microscopic view of the Al_2O_3 seed and the $Ag+Al_2O_3$ seed are shown in Fig. 5 (a, b).



(a) *(b)*

FIG. 5. SEM images of (a) Al_2O_3 rod and (b) Ag+ Al_2O

To test the adsorption performance of the ^{125}I carrying media, each rod is contacted with a ^{125}I solution of 5 mCi (185 MBq) at different solution volumes and pHs. As shown in Fig. 6(a), the adsorption capacity from the contact of a rod with a 5 mCi/50 $\mu\ell$ solution is more than 95% after 4 hours. At the same condition, the adsorption capacity of the bare silver rod is below 20%. The adsorption rate showed that the $Ag+Al_2O_3$ rod is more efficient than the pure Al_2O_3 rod and the bare silver rod. Fig. 6(b) shows the adsorption capacity of the $Ag+Al_2O_3$ rod with respect to the concentration of a solution. The equilibration times for the ^{125}I adsorption on the $Ag+Al_2O_3$ rods are 4, 8, and 24 hours when the seeds are exposed to 50, 100, and 200 $\mu\ell$ solutions, respectively. No solution pH changes (9~12) are observed after the equilibration.

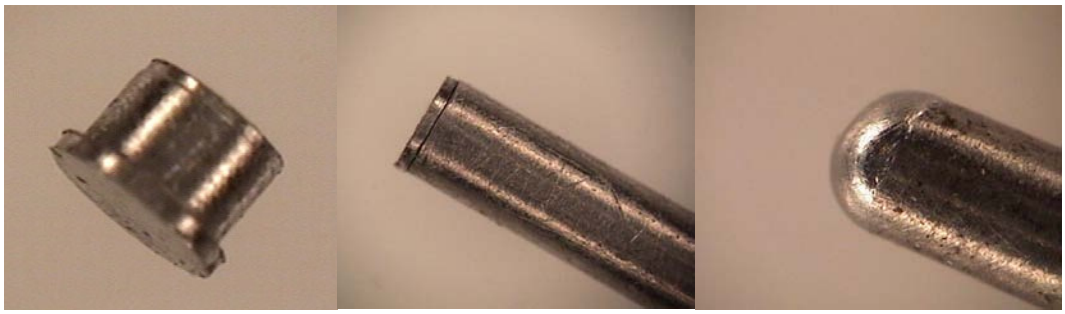


(a) *(b)*

FIG. 6. Adsorption ratio according to (a) time and (b) concentration ratio

3.2. Development of welding technique

In order to develop the welding technique for the fabrication of the ^{125}I seeds, a precise welding assembly system that adopts a rotating welder built. Two welding methods for sealing the iodine-absorbed media were tested. For the first technique, titanium capsules are designed and manufactured by using a precision abrasive machining as shown in Fig. 7(a, b). Dummy capsules are fabricated manually to demonstrate the feasibility of the proposed design as shown in Fig. 7(c).



(a) *(b)* *(c)*

FIG. 7. Photographs of cap and tube; (a) titanium cap, (b) assembly of capsule before welding, (c) dummy capsule

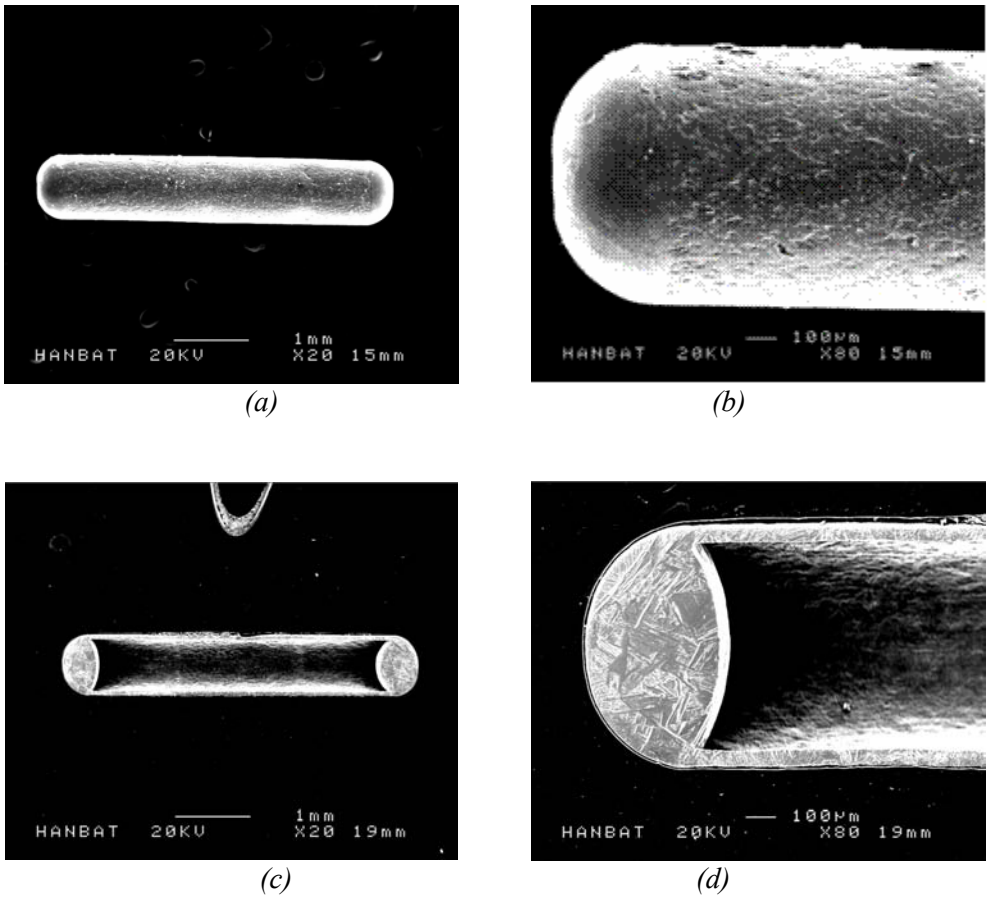
Since the cap is too small to fabricate precisely, it requires a relatively high cost to produce the capsule elements. Hence, a direct welding method, which irradiates the laser beam directly to the edge of a titanium tube, is developed.

When the edge of a small titanium tube is irradiated by a Nd:YAG laser beam, it is melted down to the proper length, coalesced, and sealed. Accurate control of the melting length of the tube edge is the most important parameter when producing sound sealings. The effects of the laser welding parameters on the melting length are analyzed and optimized by the Taguchi and regression analysis method. The pulse width and focal position among the welding parameters have the greatest effect on the S/N ratio of the melting length. Optimum welding conditions are obtained at 0.86 msec of the pulse width and 3.19 ~ 3.35 mm of the focal position in the scope of the experiments. The optimum welding conditions are finally derived as shown in Table III.

To confirm the estimated results, repeated experiments are performed with 3.3 mm of the focal position, and the average melting length is measured as 1.33 mm which agrees well with the estimated value. Fig. 8 shows the surface appearance and the cross-section of the titanium tube sealed with the 3.3 mm of the focal position and the other conditions as described in Table III. Photomicrographs show a good surface appearance with no pores and defects.

TABLE III. OPTIMIZED WELDING CONDITIONS FOR THIS DIRECT METHOD

Parameter	Nozzle type	Rotating speed	Tilt angle	Pumping voltage	Pulse frequency	Pulse width	Focal position	Welding time
Optimized condition	B	75 rpm	15 ^{o}	242 V	70 Hz	0.86 ms	3.27 mm	2 sec



(a) *(b)*

(c) *(d)*

FIG. 8. SEM photomicrographs showing the surface appearance and cross-section microstructure of a seal-welded Ti capsule((a), (c): overall, (c),(d): tube end

4. ^{169}Yb INDUSTRIAL SEALED SOURCE

Ytterbium (^{169}Yb) is in high demand in the industrial and medical fields because of its relative low effective energy, i.e. 200 keV compared to 400 keV of ^{192}Ir, and good performance with a non-beta emission. Small radiation field of ^{169}Yb provides an improved safety and cost reduction, which is associated with the number of persons required to maintain a radiographic area. Therefore the ^{169}Yb source could be a good replacement for the iridium source for radiography of thin steel structures.

It is advisable that the ^{169}Yb source has approximately 185 ~ 370 GBq (5 ~ 10 Ci) activities as a point source to apply to a radiographic industrial NDT application. The carrier free ^{169}Yb can be obtained by using cyclotron with the ^{169}Tm (d,2n) ^{169}Yb reaction. However, this method has limitations due to an unstable supply and limited production quantity. Hence, a neutron irradiation is more suitable for a stable supply and mass production of ^{169}Yb. The production scheme of ^{169}Yb by a neutron irradiation uses the ^{168}Yb (n, γ) ^{169}Yb reaction. Since a natural ytterbium target contains ^{168}Yb at only 0.13%, an enriched ytterbium target is required in order to attain a proper specific activity.

4.1. Preparation of Yb core and target

^{169}Yb source developed at KAERI is a cylindrical pellet. To fabricate the cylindrical pellet of Yb_2O_3 at the dimension of 1mm in diameter and 1mm in length, compacting equipment and a die have been developed (Fig.9). Various experiments are conducted to obtain the optimal compacting and sintering conditions. The effect of a compaction force on the density of the pellet are determined by measuring the densities of the pellets after a compaction with a force of 40 – 100 kg_f, and then after sintering the pellets for five hours at 1150^{o}C. As shown in Fig 10(a), the density of the pellets increases as the compaction force is raised. It is obvious that the density will be higher when the force is larger than 100 kg_f. However, the experiments were only performed with a force less than 100 kg_f because the hard metal employed had an allowable mechanical strength of less than 100 kg_f.

To investigate the soundness of a pellet after a compaction and sintering at various sintering conditions, Micro-Vickers hardness of the pellets is measured. As shown in Table IV, total 27 cases are studied and then the hardness of the sintered pellets is measured. As shown in Fig. 10(b), the best hardness could be achieved at the conditions of a 100 kg_f compaction and an 1150^{o}C sintering for three hours.

From a thorough analysis for the specific activity of the pellet after neutron irradiation at the HANARO, it is concluded that the pellet with the dimension of 1mm diameter and 1mm length and a density of 6.0 mg/mm^3 could have approximately a 7 Ci (259 GBq) of activity. This density can be achieved by a compaction with approximately 100 kg_f as shown in the Fig 10(a).

TABLE IV. CONDITIONS FOR COMPACTING AND SINTERING (UNDERLINE: OPTIMAL CONDITION)

Compacting Force, kg_f	Sintering Temperature, ^{o}C	Sintering Time, hr
50	900	1
80	1000	3
100	1150	5

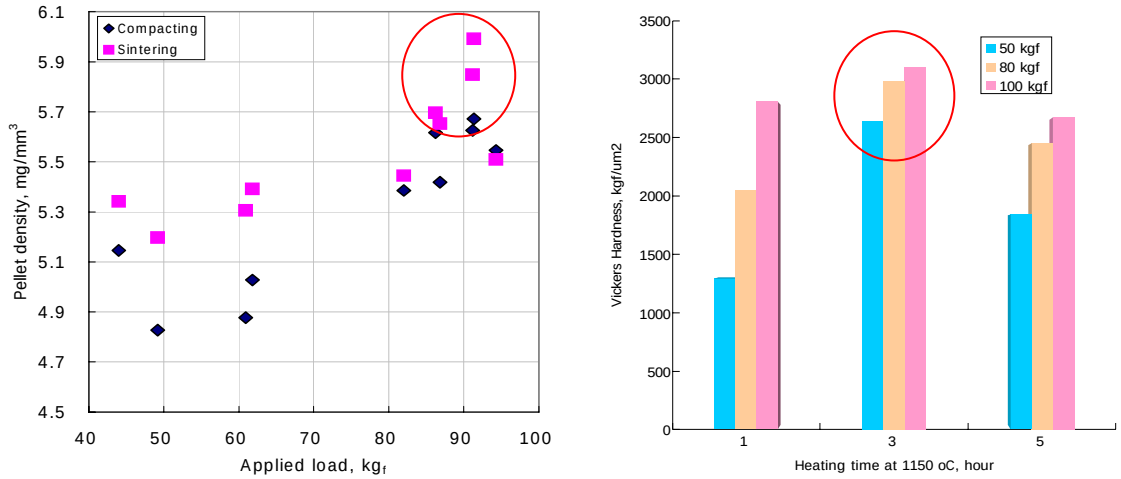
TABLE V. OPTIMAL LASER WELDING CONDITIONS FOR SEALING THE TARGET CAPSULE

Speed	Input volt.	Frequency	Pulse width	Shielding
80 mm/min	200 V	20 Hz	2 msec	15 l/min, Ar
Pulsed Nd-YAG laser welding, Specimen rotating 450 ^{o}				

Irradiation target of the ^{169}Yb low energy sealed source is developed by a welding of a pure aluminum cap and capsule in which an Yb pellet is contained. The welding for the aluminum capsule is performed by using an Nd-YAG pulse laser. To determine the proper welding conditions, the cross-section of the welded part of the capsule is inspected after a welding at various conditions. At the conditions summarized in Table V, the optimal welding is achieved with the cross-section shown in Fig. 11.

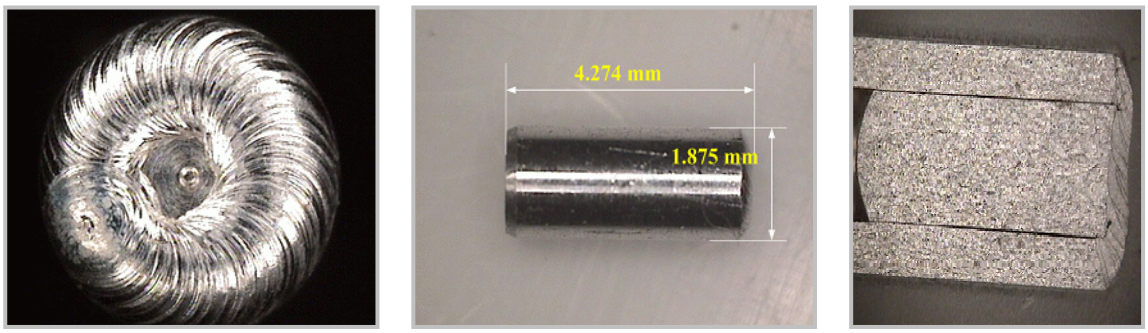
(a) (b) (c)

FIG. 9. Photographs of (a) compacting equipment, (b) die, and (c) Yb core after compacting and sintering



(a) (b)

FIG. 10. (a) Green densities of the Yb_2O_3 pellet according to various applied forces, (b) Micro-Vickers hardness of Yb_2O_3 pellet with respect to sintering time



(a) (b) (c)

FIG. 11. (a) Welding bead shape of target capsule, (b) photograph of target after welding and (c) microscopic view of welding cross-section.

4.2. Design of source assembly

Two different capsule assemblies are designed and fabricated to accommodate the ^{169}Yb source. In both cases, ^{169}Yb sources are enclosed in ^{192}Ir source capsules in order to use the NDT projectors for the ^{192}Ir source assembly in the market. As shown in Fig. 12(a), connector, pig tail, and capsule are the same as the ^{192}Ir source assembly except that the ^{169}Yb source is installed by a left-handed screw fastening in the capsule. In addition, the pig tail is fabricated at 3.8 mm longer than the ^{192}Ir source assembly because the ^{169}Yb source capsule is produced at 3.8 mm shorter than that of ^{192}Ir. The second assembly as shown in Fig. 12(b) is designed to reduce the radiation shielding of the source capsule in which the ^{169}Yb source is installed as it is welded inside the aluminum capsule. Fig. 13 shows the second source assembly produced at KAERI.

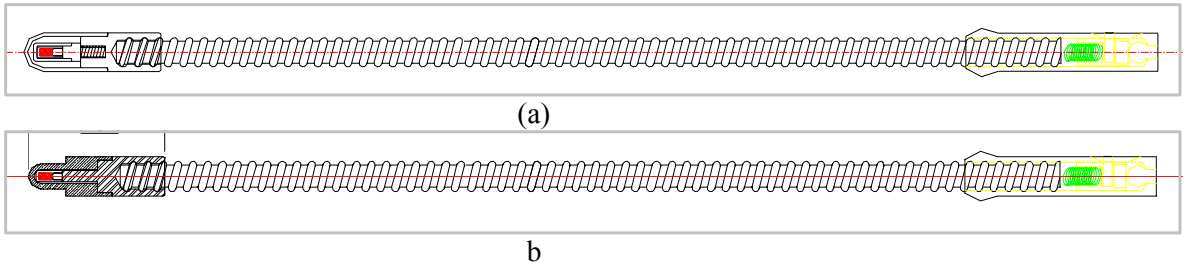


FIG. 12. Drawing of Yb-169 source holder; (a) Type I, (b) Type II

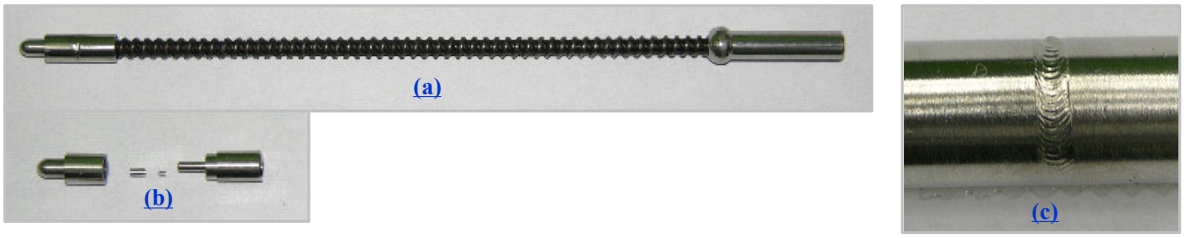


FIG. 13. Photographs of type II source holder; (a) Dummy assembly, (b) elements of source part, (c) welding area of source capsule

The developed source assembly is tested for its radiographic performance. The test results are compared with those from the ^{192}Ir and soft X ray radiography. As shown in Fig. 14, the radiography photo by the ^{169}Yb source assembly has a better resolution than the ^{192}Ir source in detection for welding defects on the carbon steel with less than a 4mm thickness.

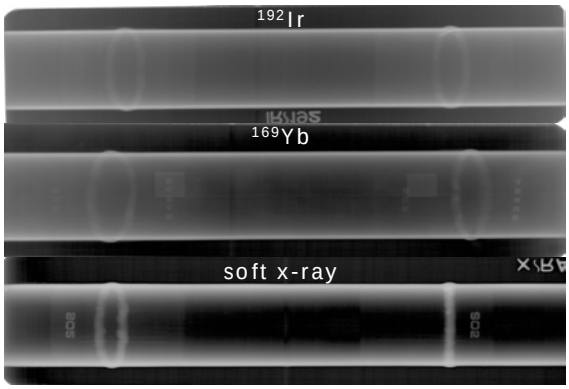


FIG. 14. Performance test results for comparing ^{192}Ir and soft X ray

5. CONCLUSIONS

As an ongoing effort to develop radiation sealed sources for medical and industrial applications, KAERI has developed ^{192}Ir sealed sources for a high dose rate brachytherapy. From a series of systematic tests, this source shows satisfactory properties from radiological, medical, and mechanical aspects and can be applied to medical treatments. And these source capsules also passed safety tests that followed ISO2919 standard.

For the case of ^{125}I, research is still in progress to develop various adsorption methods and sealing techniques. Preliminary studies show that a ceramic rod treated with silver nanoparticles has a good loading capacity for iodine. Two types of sealing technologies, a welding technique with a precision cap elements and a direct tube welding technique, have been developed. The effects of the Nd:YAG laser welding parameters on the melting of the titanium tube are analyzed by the Taguchi and regression analysis method. The optimum welding conditions for the sealing of the titanium tube ends are obtained.

^{169}Yb industrial NDT sealed sources are developed by using Yb_2O_3 pellets as the target and demonstrated for their performance. To produce the pellets, optimal compacting and sintering conditions are determined experimentally. Source holders for ^{169}Yb are designed and fabricated. After assembling an active source produced from HANARO with the developed source holder, a demonstration experiment is performed to compare the quality of the radiographs from ^{192}Ir and soft X-rays. This demonstration study shows that the developed ^{169}Yb produces better radiographs than ^{192}Ir for the carbon steel with less than a 4mm thickness. To assure safety of source capsules, safety tests in accordance with ISO 2919 will be conducted in near future.

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PRODUCTION OF ^{125}I SEED SOURCES FOR BRACHYTHERAPY USES

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Abstract

The production of radioactive sources of ^{125}I, used mainly for the brachytherapy of prostate and ocular cancer, is a work that is being carried out in the plant of production of radioisotopes (PPR) of the Nuclear Center Racso of the IPEN. The employed methodology is based on the ^{125}I physical-chemistry adsorption at silver wires coated with palladium. In the realization of the tests, it has been considered the procedure used by India and Iran participants of this CRP. In the execution of this work, the ^{131}I radioisotope is been used simulating the ^{125}I, because in the PPR-IPEN we produce the ^{131}I. In total 50 samples were used, they were divided in ten groups. In first place with nine working groups, the optimum conditions for work for the coating of the silver wires with palladium were obtained, these being the following: simple method, employing $PdCl_2$ 0.1 m, pH of 5.5 to 6.5 and a temperature of 100°C. Later on, a series of tests were carried out to determine the appropriate parameters for the adsorption of ^{131}I in the previously treated wires, these being the following: carrier concentration of Ki = 0.03 m, time of adsorption of 6 hours, and temperature of 70°C. Finally, the percentage of ^{131}I adsorption was obtained in the silver wires tried previously with palladium chloride solution of 98.24%. The control of leachability was made, having very good results. To confirm these previously mentioned parameters, a test was made with ten pieces of silver wires, corresponding to the group 10, giving the confirmation as a result of these. Also, samples of the titanium tube have been sent for test with microplasma welding to a French company: air welding liquidates export. With these results obtained, subsequently the production of these radioactive sources will be carried out employing ^{125}I as a radioisotope.

1. INTRODUCTION

Since 1970, the radioisotope commonly used in the production of radioactive sealed sources for brachytherapy uses is the ^{125}I, that it liberates 90% of his energy in 6 months, for that it is used in tumors with degrees Gleason of [2–6], and the ^{103}Pd that liberates 90% of their energy in 2 months, is used in tumors less differentiated with Gleason [7–10]. The radiation dose liberated by the ^{125}I of 160 Gray = 16,000 mGy or rad, 59 day half-life allows easy inventory management. Low 28-32 keV X rays are easily shielded to protect personnel from exposure. Sizes of these sources are like a grain of rice (0.8 mm OD x 4.5 mm long).

One of the main applications of this type of sources is in the treatment of prostate cancer, but they also are employed in the oncological treatment of head, neck, lung, pancreas and eyes, among others. It is known that the radiation alters the DNA or RNA of the neoplasic cells, so the cells when they are going to be divided they do it in an inappropriate way and die. Due to that the cells well and moderate differentiated of the prostate cancer, generally they divide slowly, these cells can survive various months after taking the implant, for which the ^{125}I is preferred because it has a more prolonged average life, while the cells not well differentiated are divided more quickly, for which the ^{103}Pd is preferred because it has an average life more short and liberate its energy with greater velocity.

The implants can be placed in a permanent or temporary form. In permanent form, 100 seeds in the prostate gland are placed approximately. Thus they will release their radiation in low doses during various months and the seeds once exhausted, they remain in the gland permanently.

The brachytherapy in the processing of the prostate cancer located, is an effective therapeutic alternative, with slight mortality, economic, with a short period of recovery post implanted that permits all patients the earliest incorporation to their habitual activities.

There are different methods of production of these sources, but in all these cases, the radioactive source is found hermetically sealed in capsules of titanium with dimensions of 0.8 mm diameter x 4.5 mm long. Seed sources before their employment in brachytherapy, are tested by a series of rigorous quality controls that guarantee their quality.

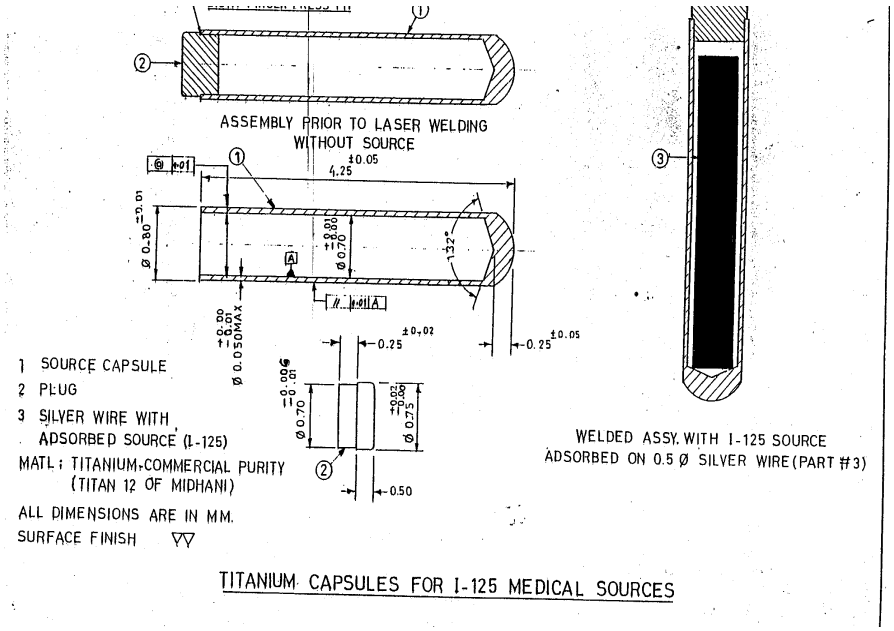

FIG. 1. Titanium capsules for ^{125}I medical sources

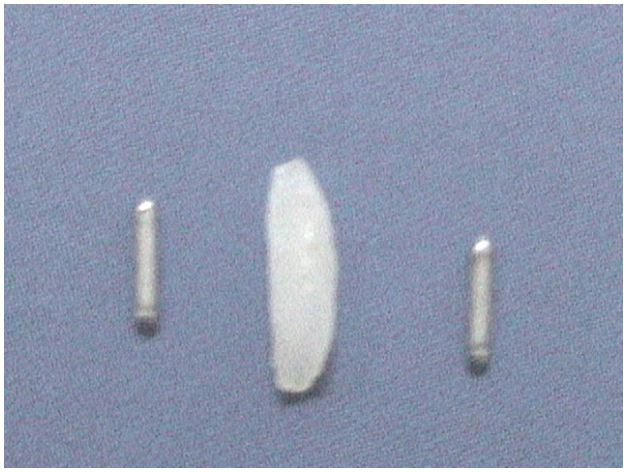

FIG. 2. The sizes of these sources are like a grain of rice (0.8 mm OD x 4.5 mm long)

2. MATERIAL

- Silver wire of 0.5 mm diameter of 99.9985% purity obtained from Alpha Aesar.
- Na ^{131}I, produced in the PPR- IPEN, Peru.
- Palladium chloride (II), with 99.999% purity, obtained from Alpha Aesar.
- The other required reagents were acquired from reputed commercial suppliers.
- The radioisotope calibrator to determined dose was the CAPINTEC CR15, with actually calibration.

3. METHODS

The method employed for the production of the ^{131}I seed source, was the physical-chemical adsorption. In this method, is very important to consider the parameters of concentration of the

reagents, for such reason, we worked with different concentrations of $PdCl_2$, and KI as a carrier, and with different times of adsorption of the ^{131}I, too.

4. EXPERIMENTAL

4.1. Pre-treatment of the silver wires

- Cut the silver wires in 20 pieces of 4 mm.
- Leave them soaking with acetone (10 mL) for 20 minute.
- Wash them with bi-distilled hot water (3 times with 10mL) and then water cold (3 times with 10mL).
- Treat the wires with HCl 3M (3 times with 5mL) and subsequently to wash them with acetone (2 times with 3mL), then with bi-distilled hot water (2 times with 10 mL) and then cold water (one time with 10mL).
- Dry the silver wires, under IR lamp (1 hour).
- Separate them in 2 groups of 10 wires each one.
- Weigh each group to obtain the weight average of the wires.
- Keep them in totally clean vials and to seal them.
- Weight average for wire of the Group 1: 0.00771 gr.
- Weight average for wire of the Group 2: 0.00752 gr.

The same methodology was carried out for 20 new wires, separated in two groups, being obtained the following weights averages:

- Weight average for wire of the Group 3: 0.00775 gr.
- Weight average for wire of the Group 4: 0.00776 gr.

FIG. 3. Pre-treated silver wires

4.2. Coating of the silver wires with palladium

Two methods employed: a simple and complex method.

4.2.1. Simple Method: the wires of the Group 1 were employed

- Submerge the wires in a solution of palladium chloride 0.03M (0,0532 gr. $PdCl_2$ /10 mL H_2O), and to pH 5.5 - 6.5. Heat the solution to dissolve.
- Heat the solution gradually up to 100°C, to maintain it to this temperature for 20 minute and then cool to environment temperature.

- Wash the wires with acetone (2 times with 3mL) and then with hot bi-distilled water (5 times with 10mL).
- Dry off to obtain a constant weight with IR lamp by one hour.
- Weigh the 10 wires recovered with chloride of palladium, to obtain the average weight. The weight of palladium covered by wire was 0.00048 g.
- Store the wires in a vial sealed and lettered until they are utilized. This group will be denominated Group 1-R

Observation: It can be observed that the wires remain coated in uniform form of dark color (leaden). Due to that good results were obtained, it would be processed subsequently to carry out new tests, but modifying the concentration of chloride of palladium.

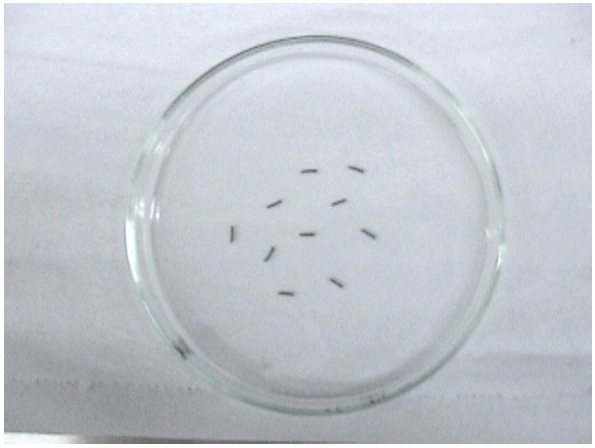

FIG. 4. Silver wires coated with palladium employing the Simple Method

4.2.2. Complex method: the wires of the Group 2 were employed

- Submerge the wires in complex composition of silver bathroom and palladium chloride of 0.1 mol/L (0.1773 g $PdCl_2$/10 mL H_2O) previously heated approx. 5 minute till its complete dissolution, formaldehyde 2 mol/L, nitric acid 1 mol/L, formic acid 0.4 mol/L. All as a complex mixture previously prepared
- Heat the solution at 30°C in a water bath and maintain it to this temperature for 35 min. And then to cool to environment temperature.
- Wash the wires with acetone (3 times with 3 mL) and then with hot bi-distilled water (5 times with 10mL).
- Let them dry off to obtain a constant weight with IR lamp by one hour.
- Weigh the 10 recovered wires, to obtain an average weight.

4.2.3. The average weight of palladium coated by wire was of 0.00057 g.

Store the wires in a vial sealed and lettered until they are utilized. To this group will be denominated Group 2R

Observation: It can be observed that the wires are not uniformly coated and detachment of palladium can be seen when they are manipulated. There are some stuck and when one tries to separate them the layer of palladium is lost. The same methodology for a group of 10 wires was carried out, employing $PdCl_2$ 0.03 M (Group 3R), being obtained similar results.

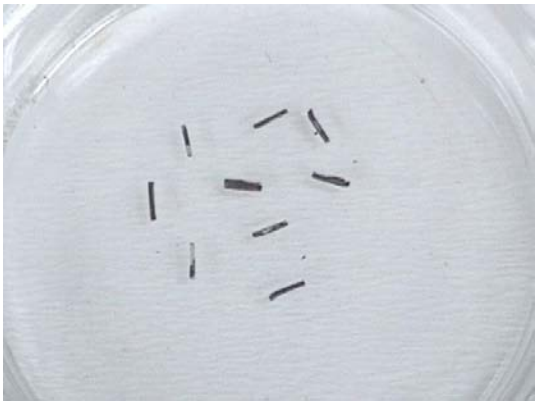

FIG. 5. Silver wires coated with palladium employing the Complex method

4.2.4. Simple method: the wires of the Group 4 were employed

It has been employed two solutions of different concentrations of $PdCl_2$, of 0.1 M and 0.03 M, to obtain the optimum concentration. They were separated in two groups:

- Group 4 A: 5 wires employing $PdCl_2$, of 0.1 M
- Group 4 B: 5 wires employing $PdCl_2$, of 0.03 M

Average weight obtained:

- Group 4 A: 0.00046 g of $PdCl_2$
- Group 4 B: 0.00038 g of $PdCl_2$

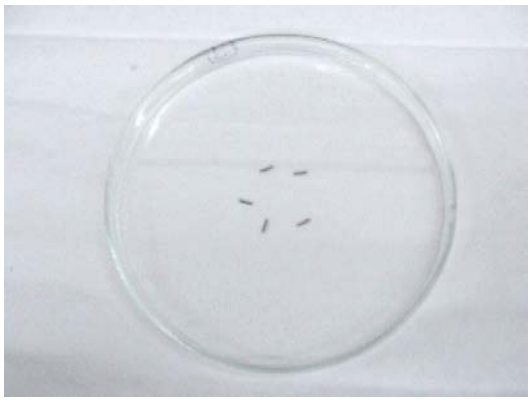

FIG. 6. Silver wires coated with palladium employing the simple method

4.2.5. Adsorption of ^{131}I on the coated silver wires with palladium

- Submerge each one of the groups of the silver wires in to solution of 80 μL of $Na^{131}I$ (≈ 4 mCi approx. 50 mCi/mL) and 200 μL of KI as carrier.
- Take the adsorption to 70°C for 1 to 6 hours.
- After every hour, to move away the wires with supreme care, by means of to clip of tip, wash them with cold bi-distilled water and proceed to calibrate them each one.
- Proceed it in the same way from the steps 2 and 3, until completing the 6 hours.

Observation:

- In the first place, the work was carried out during a time of 3 hours, to determine the adequate concentration of KI as carrier, dividing the work in 8 groups.
- 4 Groups were worked with KI 0.1 M and the 4 remaining with KI 0.03 M (Table I)

Groups formed:

- G1: 5 wires of 1-R employing KI 0.03 M.
- G2: 5 wires of 1-R employing KI 0.1 M.
- G3: 5 wires of 2-R employing KI 0.03 M.
- G4: 5 wires of 2-R employing KI 0.1 M.
- G5: 5 wires of 3-R employing KI 0.03 M.
- G6: 5 wires of 3-R employing KI 0.1 M.
- G7: 2 wires of 4-A employing KI 0.03 M.
- G8: 3 wires of 4-A employing KI 0.1 M.

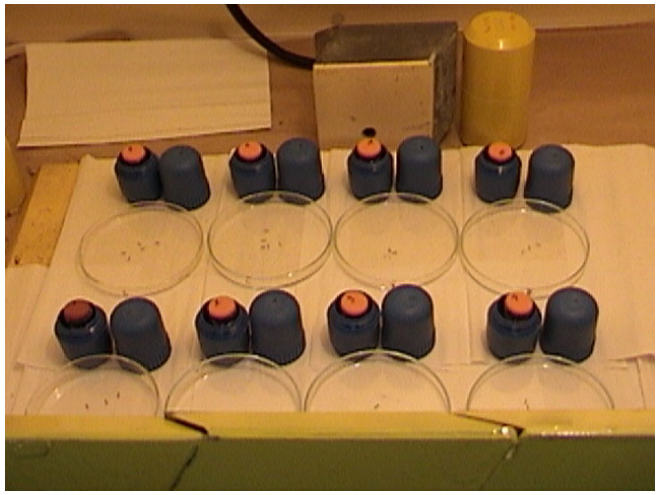

FIG. 7. Silver wires coated with palladium after the process of adsorption of ^{131}I

Finally, based on the results obtained in the Tables II and III, we worked with concentration of KI 0.03M, using the 5 wires of the Group 4-B, which was denominated G9, giving them a time of adsorption of 6 hours (Table V)

4.2.6. Studies of the leachability of the ^{131}I activity adsorbed on silver wires

- Submerge each group of the wires (G1 – G10) approximately 7 mL of bi-distilled water (temperature ambient of 22 °C). Leave them for 72 hours.
- Later on, measure the activity of the water to determine the leachability.(Table IV)

4.2.7. Verification studies of obtained optimum parameters

They were carried out tests with 10 pieces of silver wires, following the same methodology used with the Group 07 and 09. The obtained results were similar to those of the Group 9. (Tables IV&V).

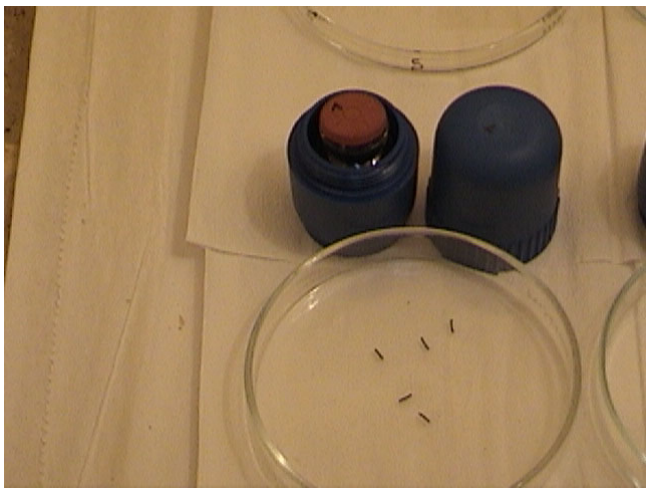

FIG. 8. Group G9 of silver wires coated with palladium after the process of adsorption of ^{131}I during 6 hours

4.3. Tests of the capsules of titanium

4.3.1 Cutting of the capsules

The cutting of the titanium capsules to the dimensions to be used in these sources was made with small diamond disks, for this aim was designed and built a mechanical system of cutting. This machine was fabricated by IPEN-Peru (Fig. 9).

Observation: The borders don't present fins, neither sinking of the material.

4.3.2 Welding

Tig welding: It was performed a test using tungsten inert gas (TIG), one ampere of current, and electrode of tungsten-thorium.

Observation: Apparently good results were obtained, but when this one was seen in the magnifying glass, imperfections were observed.

Microplasma welding

It was sent to a foreign company (AIR WELDING LIQUIDATES EXPORT), to make some tests by means of the microplasma technique.

Observation:

The visual aspect of the welding is good, however a problem is visualized in the magnifying glass. When looking with a magnifying glass, the welding area present always a very small hole, originated by the gases that are inside the tube which increase the pressure when warming during the trial of welding and they spread to escape, what for the tube is not tightly sealing.

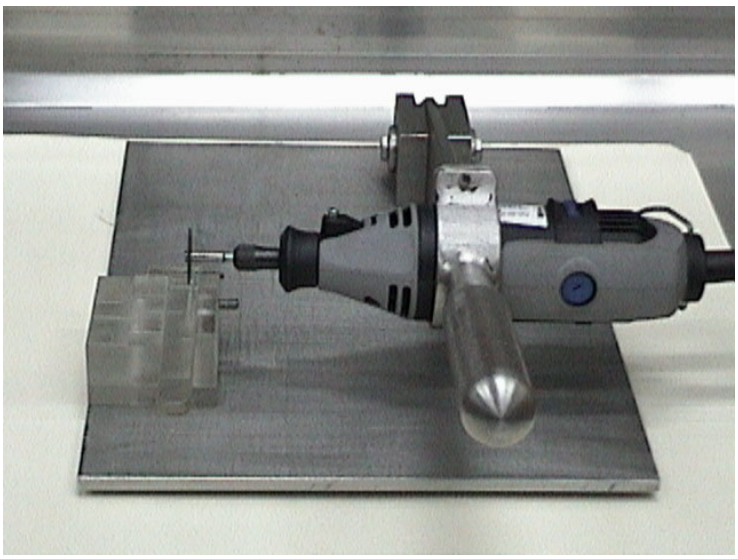

FIG. 9. Cutting machine fabricated by IPEN-PERU

5. RESULTS

Results of experiments are presented in the following tables.

TABLE I. ACTIVITY PERCENTAGE ADSORBED EMPLOYING SIMPLE METHOD AND COMPLEX OF COATING OF PALLADIUM

Method	Simple
Group	*% activity ads.*
G1	72.2 2
G2	68.8 8
G7	75.0 0
G8	61.3 3

Method	Complex
Group	*% activity ads.*
G3	46.9
G4	52.1
G5	74.6
G6	35.0

TABLE II. PERCENTAGE OF ACTIVITY ADSORBED AT DIFFERENT CONCENTRATIONS OF KI AS CARRIER.

KI 0.1 M	
Group	*% activity ads.*
G2	68.8
G4	52.1
G6	35.0
G8	61.3

KI 0.03M	
Group	*% activity ads.*
G1	72.2
G3	46.9
G5	74.6
G7	75.0

TABLE III. FINAL DATA OF THE TESTS CARRIED OUT

GROUP	COATINGOF SILVER WIRES WITH PALLADIUM				ADSORPTION of ^{131}I		
	SIMPLE METHOD		COMPLEX METHOD		KI 0.1 M	KI 0.03 M	%
	$PdCl_2$ 0.1 M	$PdCl_2$ 0.03 M	$PdCl_2$ 0.1 M	$PdCl_2$ 0.03 M			
G1		X				X	72.2
G2		X			X		68.8
G3			X			X	46.9
G4			X		X		52.1
G5				X		X	74.6
G6				X	X		35.1
G7	X					X	75.1
G8	X				X		61.4

TABLE IV. ACTIVITY OF THE ^{131}I FOR STUDY OF LEACHABILITY

Group	Activity of water (μCi)
G1	0
G2	0
G3	10
G4	0
G5	10
G6	0
G7	0
G8	0
G9	10
G10	10

A maximum percent of leachability was obtained of 0.6% and a minimum of 0%, being the average of the 10 employed groups 0.2%

TABLE V. PERCENTAGE OF ADSORPTION OF ^{131}I USING WIRE COATED OF PALLADIUM AND KI 0.03 M AS CARRIER VERSUS TIME.

Hour	% activity adsorption.
0	0
1	53.0
3	78.3
6	98.2

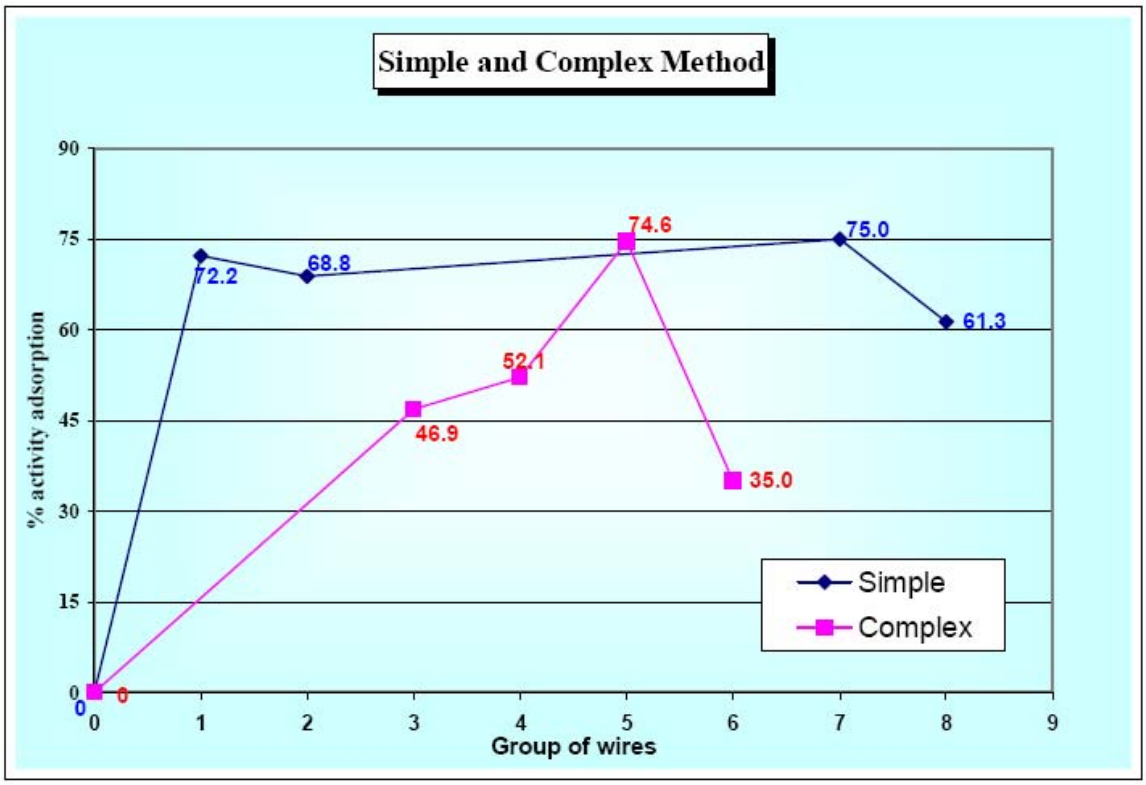

FIG. 10. Tests were carried out to volume 80 μL of ^{131}I and 200 μL of KI as carrier using the simple and complex methods. The adsorption was carried out 70°C

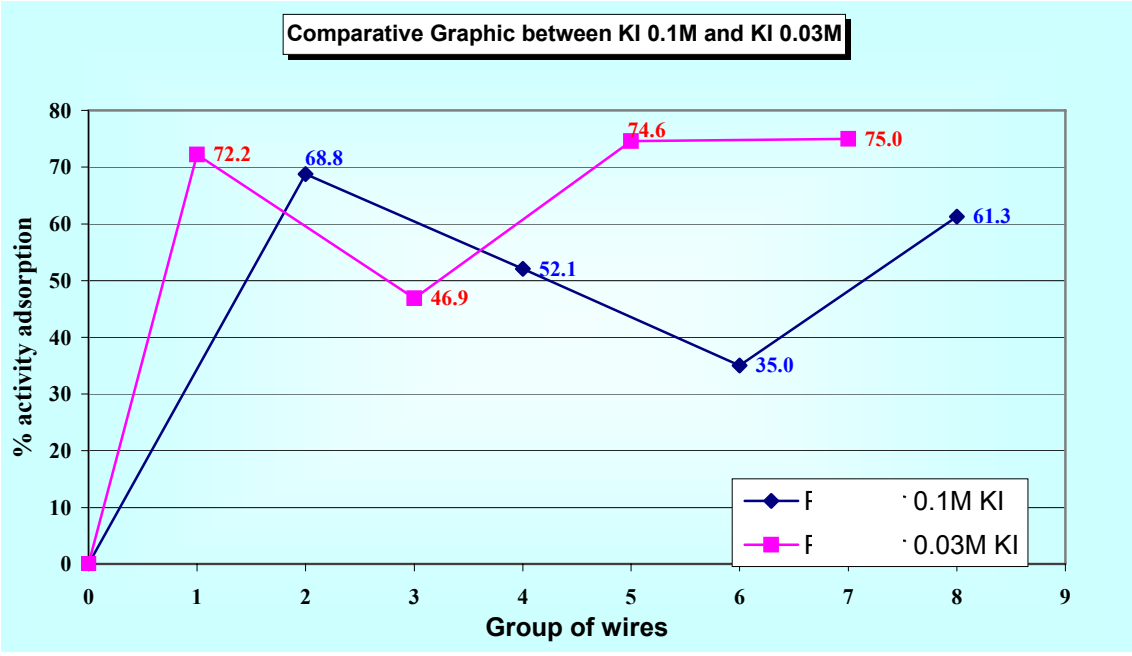

FIG. 11. Tests were carried out to a volume 80 μL of ^{131}I and 200 μL of KI as carrier. The adsorption was carried out 70°C

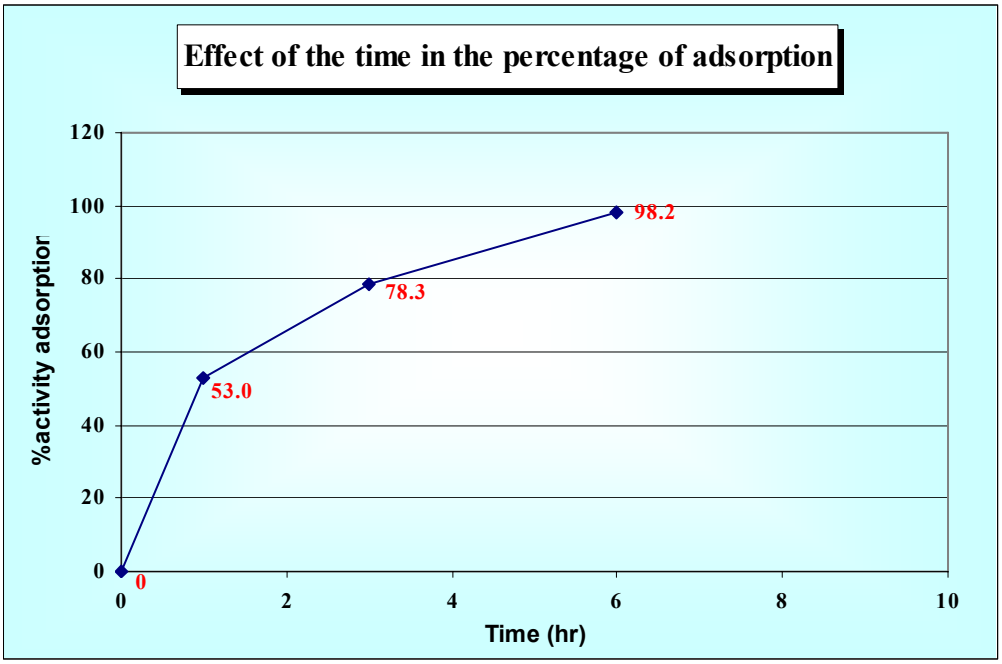

FIG. 12. Tests were carried out using the simple Method with $PdCl_2$ 0.1M and in the part of the adsorption with a volume of 80 μL of ^{131}I and 200 μL of KI 0.03 M as carrier, maintaining it to a temperature of 70°C for a lapse of 6 hours

6. CONCLUSIONS

The physical-chemistry adsorption method of ^{131}I on coated wires of silver with palladium is simple, effective and over all that in a single process can be produced a large batch of radioactive sealed sources.

It is very important to consider several factors that are critical, like the previous coating to the silver wires, the concentration and the volume of the KI as carrier, the temperature of test and the time of adsorption of ^{131}I, among others. In our case, these ideal parameters found were the following ones:

Coating of silver wires with palladium: Simple method, using a solution of $PdCl_2$ 0.1M, pH 5.5-6.5 and temperature of 100°C.

Adsorption of ^{131}I: Volume and concentration of KI as carrier = 200 μL of 0.03 M, temperature = 70°C and time = 6 hours.

Finally, it is obtained 98% of adsorption of ^{131}I and a maximum percent of leachability of 0.6% and a minimum of 0%, being the average of the 10 employed groups, 0.2%.

These previously mentioned parameters were confirmated by means of tests with ten pieces of silver wires, corresponding to the group No. 10.

Because the IPEN have not a laser welding machine, and having made tests with 2 different welding methods, TIG and microplasma to reduce the costs, we conclude that the only alternative is by means of the welding laser type.

Future work plan:

- Because they were already carried out the tests by means of the alternative techniques for their smallest cost, those of TIG and microplasma and favorable results have not been obtained, it will

be continued looking for alternatives to the financing of the purchase of a laser machine or otherwise they will be sent to abroad to seal this sources.

- Once insured the sealing of this sources by means to laser welding, will be produced sealed radioactive sources with ^{125}I, using the standardized procedure already made with ^{131}I.
- Finally, once obtained this sealed sources, it will be proceeded to make the other specific controls.

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DEVELOPMENT OF RADIOACTIVE SEALED SOURCES FOR USE IN BRACHYTHERAPY

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Abstract

The application of internal electrolysis for deposition of ^{125}I on silver was examined. These investigations were aimed at the preparation of active core of ^{125}I seed source. The cell was built of silver anode contained in Pt crucible serving as cathode. The plating solution contained $Na^{125}I$ and sodium hyroxide. The yield of ^{125}I deposition for carrier-free and carrier-containing electrolyte solution was determined. The deposition on cut into 3 mm long silver bars as also on uncut silver wires was investigated. The influence of visible light on the coating process was examined. The leachability of ^{125}I from silver wires and the activity distribution along the wires was measured. Obtained results indicate that silver wire should first be coated with ^{125}I and than cut into 3 mm long pieces. Based on this observation a device for batch-type procedure of manufacturing ^{125}I plated silver bars was designed and made. A design for simultaneous cutting and positioning of the plated with ^{125}I bars into titanium capsules is presented. The arrangement was built and tested in trials. Tests confirmed that contamination of the source capsules can be minimalized. Laser welding of the titanium capsules were performed and cross section of the welds examined by the use of optical microscope. In accordance with PN-86/J-2000 eqv ISO 2919 ISO classification testing of the designed miniature sealed source were preformed. The class obtained is C43141.

1. INTRODUCTION.

The aim of the project was implementation of the production of miniaturized radioactive sources for use in brachytherapy. It concerns the seed-type sources of ^{125}I and later also ^{103}Pd sources. Seed-type sources are used in radiotherapy of brain and prostate cancers as well as intraocular tumours (choroidal melanomas and retinoblastomas). These sources positioned in suitable assembling are used in interstitial, intracavitary and surface brachytherapy. Required activity of ^{125}I seeds used in ophthalmic applicators is about 20 mCi (740 MBq), and in other application– most often 4 -5 mCi.

Methods applied for solving the problem comprise preparation of radioactive core, 0.5 mm in diameter and 3 mm long, positioning of the core inside titanium capsule 0.8 mm in diameter and 4.5 mm long, laser welding of the capsules and performing tests for tightness and requirements in accordance with ISO 2919.

The purpose of this work comprise investigation of different parameters influencing the deposition of ^{125}I on 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.

2. EXPERIMENTAL.

The experimental techniques applied for the preparation of I-125 seed sources comprised:

- Making of radioactive source cores 3 mm long and 0.5 mm in diameter,
- Positioning of the source cores inside the titanium capsules 0.8 mm in diameter and 4.5 mm long in such a way as to avoid contamination of the capsules.
- Encapsulating of the radioactive core in titanium capsules by laser welding.

2.1. Preparation of iodine-125 seeds source core.

2.1.1. Fixing of iodine-125 on silver wires.

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

From the point of view of preparation of such 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 [1-3] was applied. This term indicates that the electrolysis of a solution occurs without the application of an external electric tension [1]. Such procedure is very simple, particularly as regards the experimental requirements which are of importance when radioactive electrolytes are used.

The deposition of iodine-125 on silver wires was studied. In these experiments, 10 ml of electrolyte solution containing 0.01 M/l NaOH and both carrier-added and carrier-free iodine-125 was used. Concentration of carrier iodine-125 in the electrolyte was 1,6 and10-4 M/l NaI. 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 1.1 GBq (30 mCi). From among different metals examined as counter electrodes, for iodine deposition platinum was chosen, as it standard potential is more positive than that of silver. The coating solution was placed in a Pt crucible which served as cathode.

Two different kinds of experiments were performed:

- In the first part of experiments the silver bars cut into 3 mm long pieces were immersed directly on the bottom of the crucible.
- In the second part, the uncut wires were immersed into the solution contacting the brim and bottom of the crucible.

During the process of deposition permanent stirring of the plating solution was not applied. Sporadically the cut bars were stirred manually. The influence of light on the deposition of ^{125}I on cut bars was investigated using an UV-Mix Lamp 300 W (Śląska Fabryka Lamp Żarowych, Poland).

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

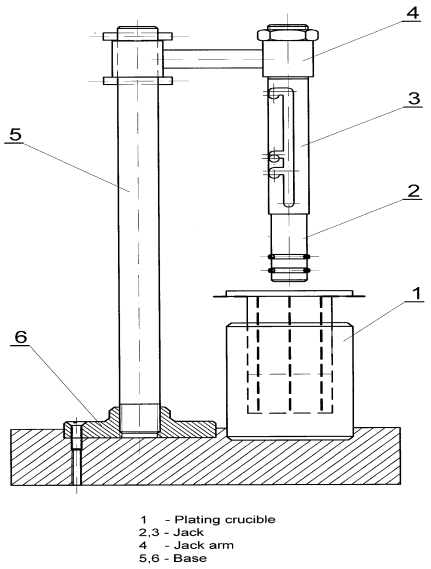
2.1.2. Design and performance of the device for fixing iodine-125 on silver bars.

Basing on the obtained results it has been decided that the technique applied for a batch type manufacturing should comprise first fixing and afterwards cutting the active silver wire into 3 mm long pieces. Following this conception, a devise for a batch-type preparation of silver bars coated with ^{125}I was designed and made. This arrangement shown in fig. 1 enables preparation of 4 active bars, each 12 mm long, in a single procedure although the number can be increased according to demands.

The plating assembly is placed in a glove box connected with radioactive fume cupboard, where the procedure of coating of silver with iodine-125 is performed (fig.2).

The plating arrangement shown in fig.3 consists of:

- Ag wire holder
- Blank holder with pressure pins
- Shielding container
- Pt crucible
- Ag wire
- Plating solution



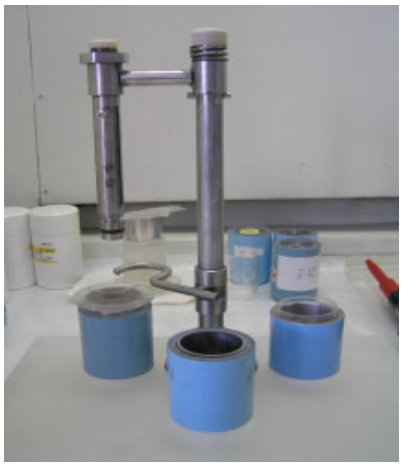


FIG. 1. Plating assembly

FIG. 2. Plating assembly stand

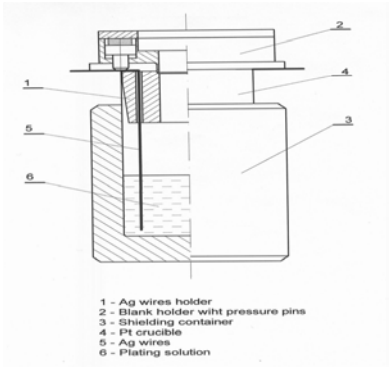


FIG. 3. Plating arrangement

The procedure of coating comprises the following steps:

- Filling the platinum crucible (4) with appropriate volume of plating solution
- Positioning the wire holder (1) on the platinum crucible (4)
- Insertion of the silver wires into the holes of the wire holder (1)
- Positioning the blank holder with pressure pins (2) on the wire holder (1)

A central hole in the blank holder and wire holder enables collection of plating solution samples for activity measurements during the coating process.

After completion of the plating process the operation should be as follows:

- Removal off the blank holder
- Insertion of the jack pilot bar into the hole of the wire holder
- Elevation of the jack pilot bar together with the wire holder to intermediate position. The silver wires are removed from the plating solution and allowed to dry up.
- Elevation of the jack pilot bar to upper position and turning arm by 90 º
- For additional protection of activated wires they can be immersed in a container with 3% solution of polystyrene in chloroethylene
- Elevation of jack pilot bar together with the wire holder to intermediate position for drying.
- Removal of the wires and positioning in the cutting device (fig.4)

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

The pieces of the silver wires, coated with iodine-125 were placed in 5 ml of water at ambient temperature for 48 hours, according to PN-ISO 9978 [4]. The radioactivity of water was measured by the use of scintillation system with the NaI(Tl) cristal and also by X-gamma spectrometer with 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.

For minimizing the abrade of iodine-125 fixed on the silver backing the protection by using layer of polystyrene lacquer was applied. The leachability of ^{125}I from silver bars coated with protective layer of polystyrene lacquer as well as unprotected silver wire was measured.

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

The prepared sources were checked for uniformity of the activity distribution. The length of the active part was 10 mm. Distribution of ^{125}I along the wire, 0.5 mm in diameter was measured using gamma counter equipped with 1 mm collimator head. The energy of 35.5 keV was measured.

2.2. Source core encapsulation and laser welding of titanium capsules.

2.2.1. Cutting and positioning of activity-coated silver bars inside titanium capsules.

This process includes simultaneous cutting and positioning of active 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.

The design for cutting and positioning of the activity-coated silver bars inside the titanium capsules is shown in fig. 4.

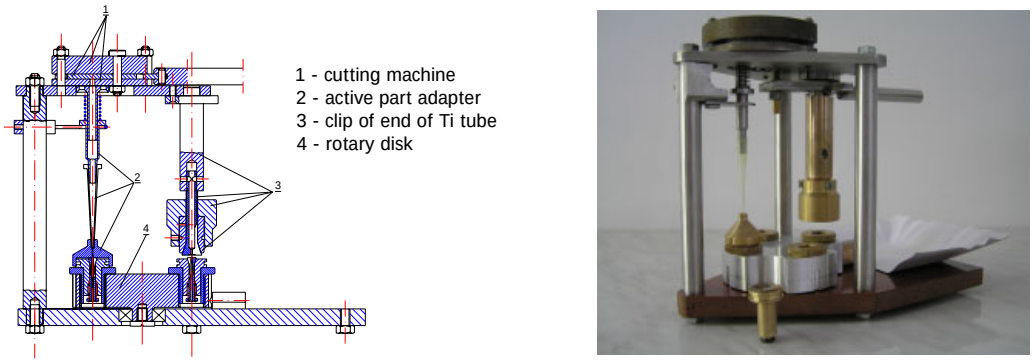


FIG. 4. Cutting and positioning device

This arrangement enables direct positioning of the radioactive silver bars in titanium tubes with previously welded bottom as well as allows one to avoid contamination of the capsules during loading which has been confirmed in few trials.

2.2.2. 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 block diagrams of this laser welding are shown in fig.5.

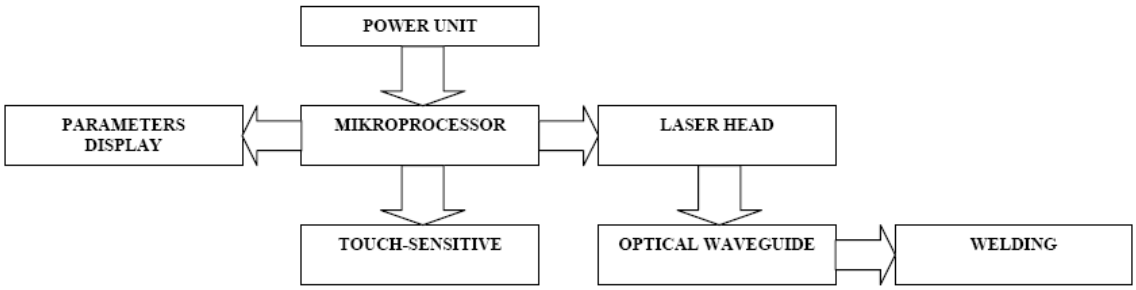


FIG. 5. The block diagrams of a pulsed Nd:YAG laser welding system CTL-1508, produced by Laserinstruments Ltd. PL

Additional accessories comprised:

- CCD camera for observation of the process on TV monitor
- A mechanical system for precise positioning of the welded element against the laser beam.

Monitor, parameters display, power unit and touch-sensitive keyboard are placed outside of the „hot cell", and microprocessor controller, laser head, optical waveguide and welding head as well as a mechanical system for precise positioning of welded element against the laser beam are positioned inside the „hot-cell". Arrangement for laser welding is shown in fig. 6.

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.

FIG. 6. Arrangement for laser welding

For precise positioning of the welded element against the laser beam, the welding head can be shifted along Z-axis, and cruciform table along X-axis and Y-axis. The stepping motor, mounted in the cruciform table enables rotation of welding element along its axis during the welding process as is shown in fig. 7.

FIG. 7. Positioning of welded element against the laser beam

Position of the welded element against the laser beam and the run of the welding process can be observed on monitor connected with CCD camera.

Four different methods for laser welding of titanium capsules were examined. In all cases the laser beam was coaxial with the welded tube. The process of welding runs without rotation perpendicular to the long axis of the welded element.

1. In the first method the open tip of titanium tube contained a Ti plug. The welding was performed by applying:

- power 5 W,
- pulse duration time 6 ms,
- pulse frequency 2 Hz.

2. In the second method studied the tip of the tube remained open. The beam parameters were:

- power 15 W,
- pulse duration time 9 ms,
- pulse frequency 2 Hz.

3. In the third method the tip of the titanium tube was squeezed oblong. The welding was performed by applying

- power 12 W
- pulse duration time 9 ms,
- pulse frequency 1 Hz.

4. In the fourth method the tip of the titanium tube was squeezed in three-armed star shape. The beam parameters were:

- power 17 W
- pulse duration time 8 ms,
- pulse frequency 3 Hz.

The cross section of the capsules were visualised by the use of optical microscope (enlargement × 50 and × 200) after polishing and etching.

3. RESULTS AND DISCUSSION

In the case of cut silver bars, they must be after coating with ^{125}I removed from the plating solution and rinsed with water and/or aceton. In spite of the small dimensions such operation can cause problems. Additional protection with a kind of lacquer may lead to sticking together after drying. Uncut wire can be after coating easily protected with e. g. polystyrene lacquer and the process of subsequent cutting and positioning in the titanium capsules can be done simultaneously.

3.1. 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 [1,2].

The effect of carrier iodide addition on yield of deposition iodine-125 on uncut silver wires is shown in fig. 8.

The results indicate that although the adsorption in the case of carrier-free solution proceeds slower, the total yield remains the same and amounts 97 %. However from point of view of fixing iodine-125 on silver, deposits of better adherence are obtained when carrier-free iodine is introduced to the electrolyte. Thinner $Ag^{125}I$ layer causes less contamination problems.

The results obtained for cut silver bars 3 mm long, immersed directly at the bottom of a Pt crucible containing carrier-free ^{125}I solution are shown in fig. 9.

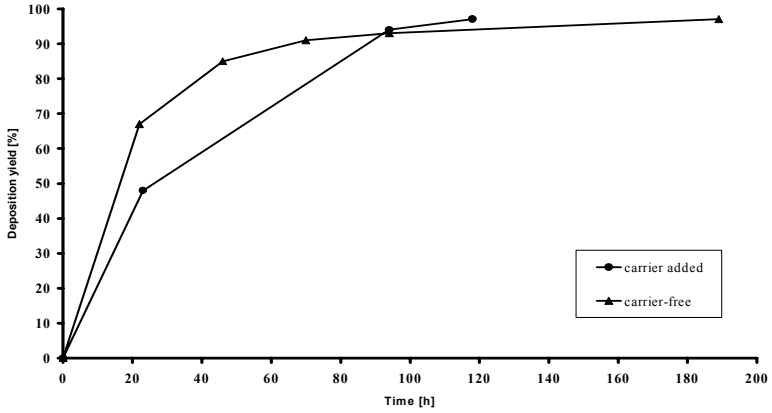


FIG. 8. Deposition yield vs. time for carrier-added and carrier-free solution

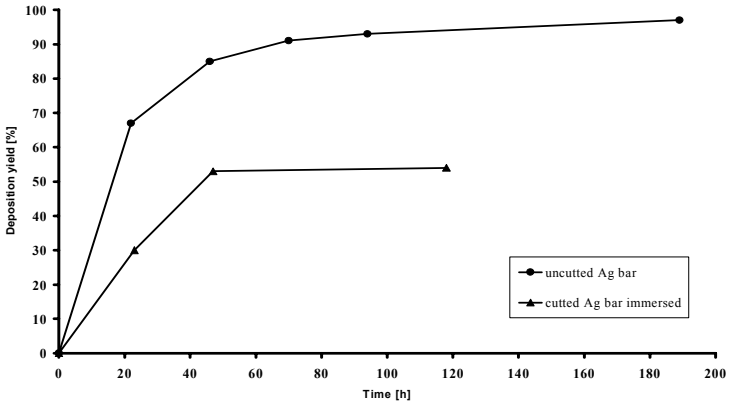


FIG. 9. Deposition yield on cut and uncut silver bars

These results indicate that the deposition yield on cut bars is considerably lower than that on the uncut ones. This can be caused by the 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 thicker silver iodide layers obtained from the carrier containing solution could cause even higher resistance and further decrease of the deposition yield. This is confirmed by the results plotted in fig. 10. The presence of the carrier reduces the deposition yield from 54 % to 29 %.

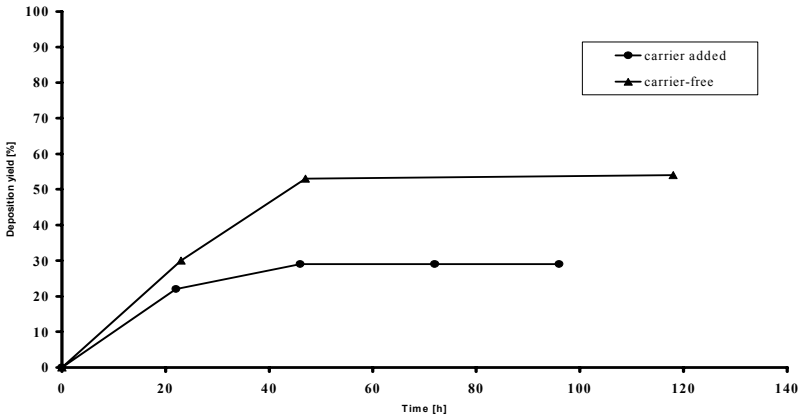


FIG. 10. Deposition yield on cut Ag bars from ^{125}I solutions carrier added and carrier free

As it follows from the literature data [5] the electric conductivity of AgI increases under the influence of light. Therefore it can be expected that the deposition yield should also increase when the solution is illuminated. It is confirmed by the results shown in fig. 11.

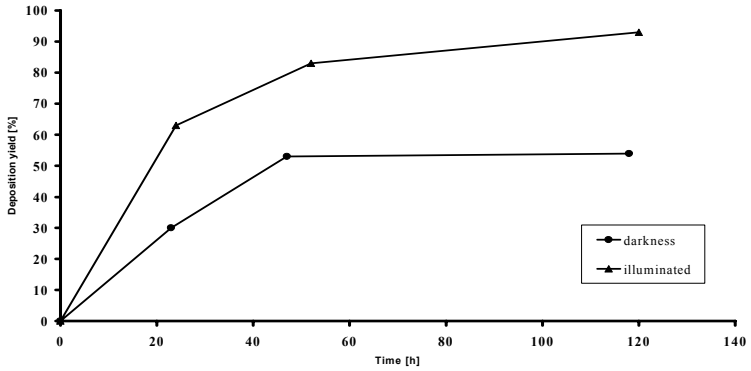


FIG. 11. Effect of illumination on deposition yield of ^{125}I on cut silver bars

Similar effect was not observed in the case of deposition on uncut bars where the electric contact between Pt crucible and bare silver wire is assured outside the plating solution (fig.12).

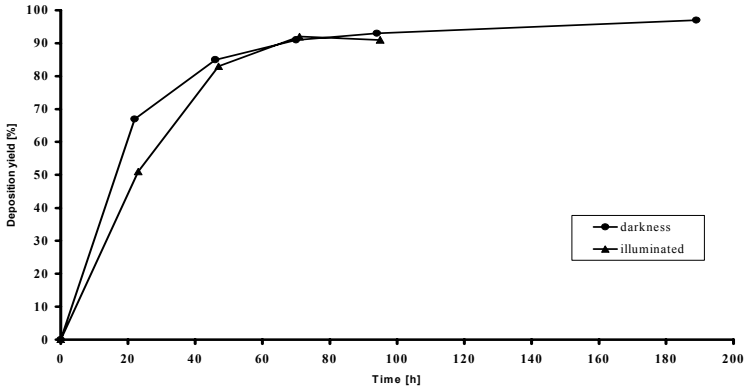


FIG. 12. Effect of illumination on deposition yield of ^{125}I on uncutted silver wire

The photoelectric effect on the conductivity of silver chloride, bromide and iodide was studied earlier [5]. 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 also 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 silver anode. On the expanded anode surface additional adsorption of radioiodine can occur. Therefore electrodeposition and adsorption can be considered as two parallel processes.

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

Results of leachability are presented in table I (Uncertainty of activity determinations ± 5%).

TABLE I. LEACHABILITY OF ^{125}I WIRE SOURCES

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

The results obtained indicate that leachability of ^{125}I from protected silver bars is very low. In case of unprotected source it is higher although still lower than other results reported [6].

3.3. Determination of activity distribution on the silver wires

The activity distributions along the protected and unprotected silver wires are shown in fig. 13 and 14 respectively.

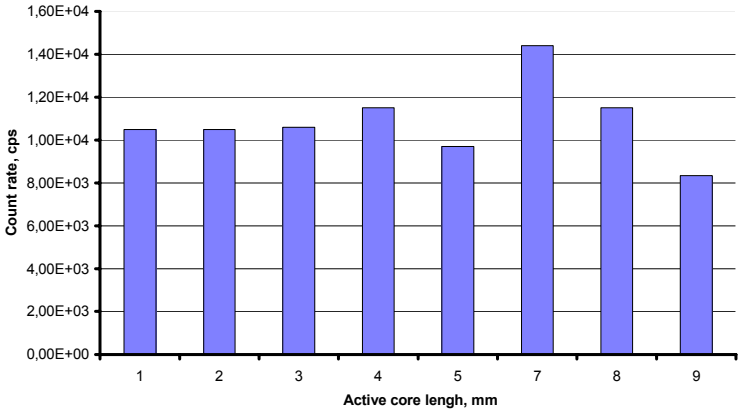


FIG. 13. Activity distribution on protected silver wire

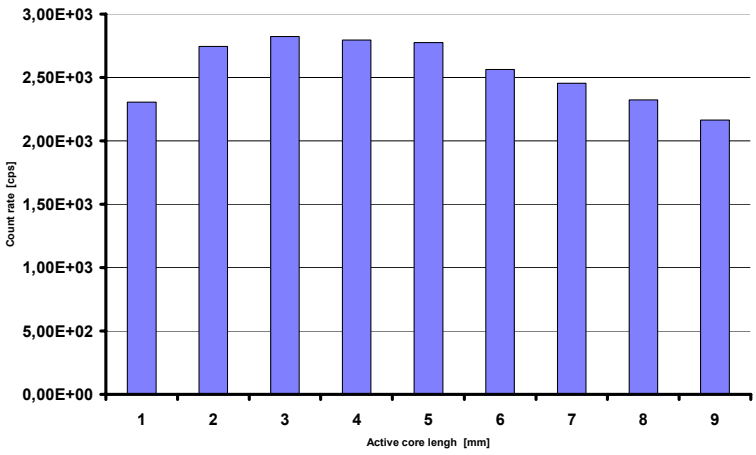


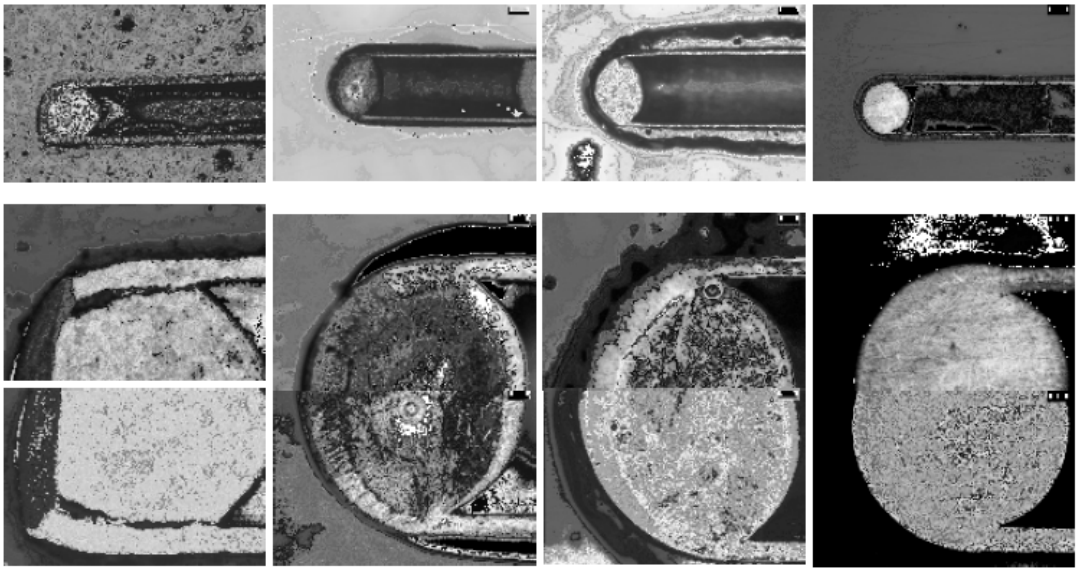
FIG. 14. Activity distribution on unprotected silver wire

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 non equivalent 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 Pt cathode. Although 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. [7] for palladium coated nickel wires.

3.4. Laser welding of titanium capsules

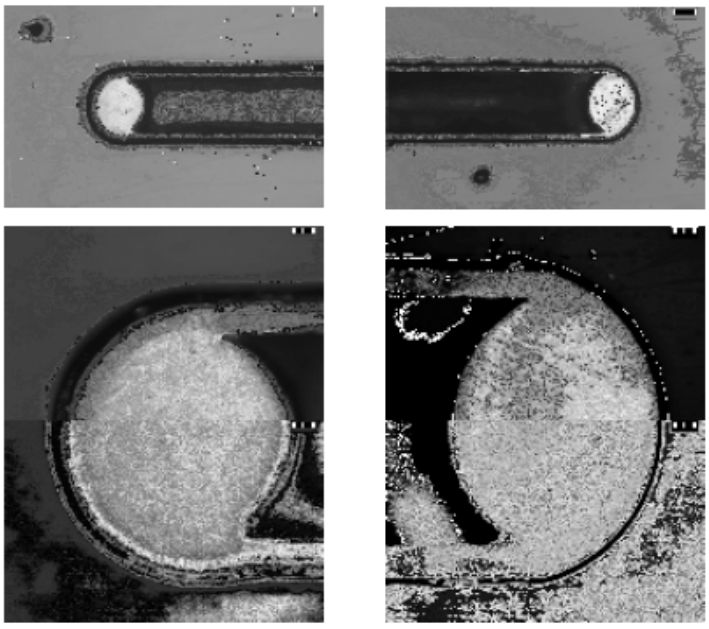
The cross sectional view of the obtained weld according to the first, second and third methods described in the experimental part is shown in fig. 15 a, b, c and must be considered as unsatisfactory.



a. b. c. d.

FIG. 15. Laser welds of titanium tubes (a) with plugged tip, (b) with open tip, (c) with squeezed tip oblong, (d) with squeezed tip in three-armed star shape

The result obtained by the use of the fourth method is pictured in fig. 15 d. The shape of the obtained weld is of good quality although the cross sectional view shows some unevenness of the melted metal. Comparison of the welds obtained in this work with these of the seeds and commercially available is shown in fig. 16.



a b

FIG. 16. Cross-section views of performed weld (a), and commercial seed (b)

The quality of both welds can be considered as similar. They have the same elliptical shape, identical structure, are free from gas cavities and from free space between the welds and internal wall of titanium tube, which were observed in the case the welds, presented earlier.

3.5. ISO classification tests.

The prototype seed sources were tested in accordance with ISO 2919. The class obtained is C 43141. This means that the seed-type sources maintained its leak tightness according to ISO 978 after testing against:

- Temperature from - 40º C (20 min.) up to +400º C (1 h),
- External pressure from 25 kPa absolute up to 2 MPa absolute,
- Impact – no test
- Vibration – 30 min 25-80 Hz at 1,5 g_n amplitude,
- Puncture – no test.

4. CONCLUSION

Among the various methods of fixing of iodine-125 on silver wire, internal electrolysis is simple and effective. The experimental condition facilitates safe handling of high amounts of radioactivity without the release of iodine-125 to the air. Using the method of short-circuited silver wires with platinum cricible outside the electrolyte solution, fixing of more than 30 mCi of ^{125}I on 3 mm long bar 0.5 mm in diameter was attained, irrespective whether the electrolyte solution contained carrier-free or carrier-added iodide solution. The yield of ^{125}I withdrawal from the solution was 98 %. However deposits of better adherence are obtained when carrier-free iodine is introduced to the electrolyte solution. Thinner $Ag^{125}I$ layer causes less contamination problems. The coated Ag bars require careful handling for encapsulation. This is assured by the designed equipment. Performed experiments in which different welding parameters were applied showed that good quality of welds can be obtained.

The class obtained for prototype seeds is C 43141. In further development of the encapsulation technique efforts will be made to obtain class C 53242, characteristic for commercially available seeds.

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PRINCIPLES DEVELOPMENT OF ^{103}Pd REACTOR PRODUCTION TECHNOLOGY AND PALLADIUM SEED CORE PRODUCTION TECHNOLOGY BY ELECTROCHEMICAL PLATING

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Abstract

A possibility of palladium-103 production by irradiation of isotopically enriched palladium-102 in the SM high-flux reactor is demonstrated. Palladium-103 specific activity at the end of irradiation was 495 Ci/g. A technique was developed for production of palladium-103 source cores by electrochemical deposition from phosphate-type electrolyte. Electrochemical deposition was carried out by plating a nickel wire (0.5 mm thick) with palladium on a single cathode and a ring-type anode cell, as well as in a cell with a flat anode allowing for simultaneous production of up to 7 cathodes. The length of the active layer on the cathode was al least 30 mm. The activity of the plated palladium-103 during production was 0.34 to 2.53 mCi. Absorption factors of palladium-103 irradiation in the cathode and the titanium capsule were determined. Measurements of palladium distribution uniformity along the full length and the lateral surface of the cathode were performed. A technique for palladium-103 electrochemical deposition from phosphate electrolytes on a nickel wire is proposed. A thorough preparation of the cathode surface is required for proper deposition of palladium.

1. INTRODUCTION

Implantation of radioactive gamma-sources is one of the most effective procedures for treatment of localized malignant tumors. Iodine-125 sources are widely and effectively used for prostate cancer treatment; these sources can be used for treatment of some other tumors, either [1, 2]. A disadvantage of iodine-125 sources is a relatively long half-life (T½ = 50 days). After implantation of the sources a patient is “radioactive” for a long period (total activity of the implants can reach several hundreds of milliCuries). Besides, in case of a source failure (unsealing) iodine-125 can be leached out from the core causing an intake of iodine-125 by the body and unjustified irradiation of other tissues and organs, i.e. thyroid gland.

An alternative to iodine-125 sources is a palladium-103 one. This radionuclide emits more “soft” photons (20-30 keV) and has shorter half-life (17 days). Metal palladium is compatible with biological tissues, is practically insoluble in water and biological liquids. The combination of these properties gives advantages of palladium-103 as compared with iodine-125; this, for example, lessens risk for a patient in case of an unforeseen source failure. However, advantages of palladium-103 can be realized if a large-scale production of these sources is set up and their price is comparable with that of iodine-125 sources. The aim of this work was as follows:

- evaluation of a possibility of palladium-103 production by irradiation in the SM reactor;
- conditioning of the sources’ cores production technique by electrochemical plating of palladium-103 onto a wire support.

2. MATERIALS

All the reagents used were of analytical grade or better, and they were used without additional purification. Palladium behavior was studied by a radioactive tracer technique using palladium-103. This radionuclide was produced by irradiation of isotopically enriched palladium-102 in the SM reactor. In a number of experiments a carrier-free isotope produced by “Cyclotron” Co., Ltd (Obninsk, Russia) was used.

3. METHODS

3.1. Production of palladium-103 in the SM reactor

Under the project trial irradiation was carried out of metal palladium enriched with palladium-102 of the following isotopic composition:

- ^{102}Pd - 89,0%
- ^{104}Pd - 3,6%
- ^{105}Pd - 4,0%
- ^{106}Pd - 2,45%
- ^{108}Pd - 0,75%
- ^{110}Pd - < 0,2%

The specimen was irradiated for 19.2 effective days in the neutron trap – the irradiation position of the SM reactor with the highest neutron flux available. Metal palladium powder was sealed in a quartz capsule, placed inside a titanium target. Palladium weight was 2 mg. This quantity was essential for production of representative experimental data after irradiation. Thermal neutron flux density inside the target volume was 1.8×10^{15} $s^{-1}cm^{-2}$, and that of epithermal neutrons was $1{,}65\times10^{15}$ $s^{-1}cm^{-2}$. Post irradiation treatment included mechanical cutting of the titanium capsule and destroying of the quarts one. Palladium was dissolved in 10 ml of the HCl and HNO_3 (1:3) mixture, followed by the activity measurement by gamma-spectrometry.

3.2. Palladium electrochemical plating

3.2.1. Procedure of electrochemical plating

Plating was carried out using electrochemical cells, presented in Figs. 1 and 2. Cell bodies were made of plexiglass. A basket-shape anode of cell 1 was made of a rhodium wire 0.4 mm in diameter. A centering hole was made in the cell bottom to prevent twisting of the support wire during the plating process caused by surface tension. The electrolyte volume was 2.8 ml.

The anodes of cell 2 were made of iridium foil. A cathode holder allowed for uniform edgewise placing of up to 7 cathodes. The electrolyte volume in that cell was 12.5 ml. Forced cell cooling was provided while electrochemical plating. The B5-50 DC power source was used for power supply. Cathodes were weighted after palladium plating using analytical balances of VLR-200 type.

After chemical treatment the support was weighted and fixed in the cathode holder. The cell was filled up with the electrolyte, the cathode was placed inside the cell and plating was proceeded using the selected mode (current, voltage, time). As a rule, palladium-103 was plated from the solution containing both natural palladium (carrier) and palladium-103 (tracer) under 10 V voltage and 1 mA current. After the plating is completed the power supply was switched off, the cathode was taken out of the cell and the cathode holder, rinsed with water, ethanol, dried under vacuum for 10 min and weighted.

3.2.2. Electrolyte composition

Three types of electrolytes were tested; they are most frequently used for precipitation of palladium for commercial and research purposes [3].

Amino-chloride electrolyte of the following composition was used:

- $[Pd(NH_3)_4]Cl_2$ (calculated for Pd) - 35 g/L

- NH_4OH (25%) 55 g/L
- NH_4Cl 18 g/L
- pH - 8,5 – 9,5

Electrolysis mode:

- Temperature (20 ± 5)°C
- Cathode current (i_k) - (1 – 2) Amp/dm^2
- The current yield is 98 %.

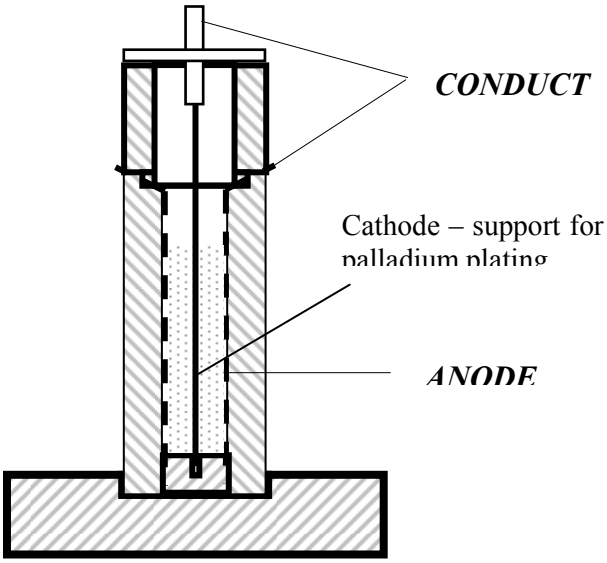

FIG. 1. Cell 1 design for electrochemical plating of palladium

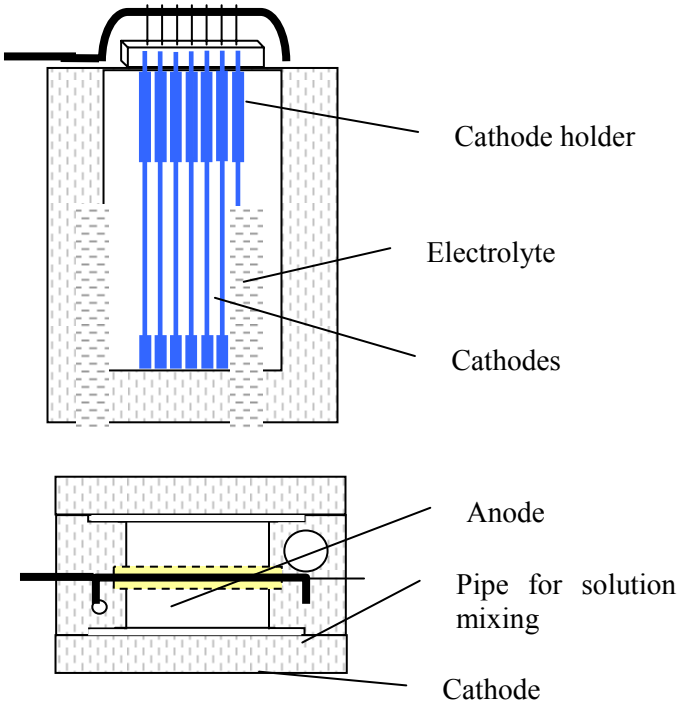

FIG. 2. Cell 2 design for electrochemical plating of palladium

Preparation of this electrolyte starts from preparation of Vockelen salt. Reaction of $H_2[PdCl_4]$ with ammonia at cooling gives hardly soluble salt of pink color via the reaction:

$$2\ H_2[PdCl_4] + 4\ NH_4OH \rightarrow [Pd(NH_3)_4][PdCl_4]\downarrow + 4\ HCl + 4\ H_2O$$

Heated in an excess of ammonia, this salt turns into a well-soluble colorless $[Pd(NH_3)_4]Cl_2$. Nitrite type electrolyte was of the following composition:

- Na_2PdCl_4 (calculated to Pd) - 10 g/L
- $NaNO_2$ - 10 g/L
- NaCl - 50 g/L.

Among the several types of phosphate electrolytes the following one was selected:

- H_2PdCl_4 (calculated to Pd) -3,5 g/L
- $(NH_4)_2HPO_4$ -20 g/L
- $Na_2HPO_4 \cdot 12\ H_2O$ -100 g/L
- NH_4Cl -25 g/L
- NH_4OH -adjusted to pH 7.

Electrolysis mode:

- T 50 °C
- i_k 0,25 Amp/dm^2
- Current yield– (90 – 95) %.

The electrolyte was prepared by mixing water solutions of its components in the following sequence: $H_2PdCl_4 \rightarrow (NH_4)_2HPO_4 \rightarrow Na_2HPO_4 \cdot 12\ H_2O \rightarrow NH_4Cl$. The solution was diluted with water till the desirable volume and NH_4OH was added to adjust pH to 7.

3.2.3. Activity counting

Activity of ^{103}Pd produced by irradiation in the SM reactor was measured using a gamma-spectrometer with an X ray detector of BDER-GA-7 k type. The activity of gamma-emitting impurity radionuclides was measured using HP Ge detector coupled with a single-board SBS PC based gamma-spectrometer.

The activity of palladium-103 plated onto cathode was measured and scanned with a single-board SBS spectrometer coupled with a BDER-GA-7 k type detector and a domestically made scanning device, controlled by PC. Detection efficacy was calibrated with a set of standard sources containing ^{241}Am, ^{113}Sn and ^{109}Cd. This set of radionuclides allows for a reliable check of the spectrometer operation and evaluation registration factor for low-energy (20 and 22.7 keV) emission of palladium-103. The geometry of samples was adjusted to avoid the measurement system overloading over 1000 pulse/sec. The error of activity counting for palladium-103 sources was less than 10% for 10 min exposition.

4. RESULTS

4.1. Production of palladium-103 using SM reactor

The reactor production of ^{103}Pd previously described in [4,5] is based on the ^{102}Pd(n,γ)^{103}Pd reaction. The scheme of nuclear transformations realized in the process of Pd-102 neutron irradiation is shown in Fig. 3.

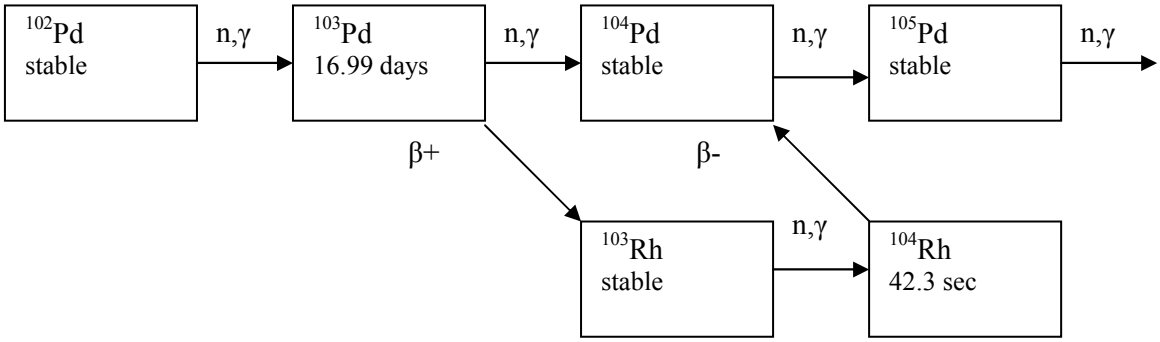

FIG. 3. ^{103}Pd production chain

Low isotopic abundance of ^{102}Pd in the natural mixture of palladium isotopes (1,02%) governs the usage of palladium highly enriched with this isotope as a target material. A small cross-section of ^{102}Pd (n,γ)^{103}Pd reaction (thermal cross-section is 3.4 barn, resonance integral 10 barn) stipulates use of high-flux reactors, like HFIR (ORNL, Oak-Ridge, USA) or SM (RIAR, Dimitrovgrad, Russia), to produce high-specific activity ^{103}Pd.

Unfortunately, the enrichment of palladium with ^{102}Pd rarely exceeds 50-80%, i.e. this material contains other palladium isotopes, like ^{108}Pd and ^{110}Pd. These isotopes produce other radioactive isotopes during neutron irradiation. For example, ^{108}Pd gives ^{109}Pd ($T_{1/2}$ = 14 h) by (n,γ) reaction, that can not be separated from ^{103}Pd chemically; another nuclear reaction ^{110}Pd (n,γ) gives ^{111}Pd ($T_{1/2}$ = 23 min) that decays to ^{111m}Ag ($T_{1/2}$ = 7.5 d); thus, a radiochemical procedure should be applied to purify ^{103}Pd from radioactive silver impurity.

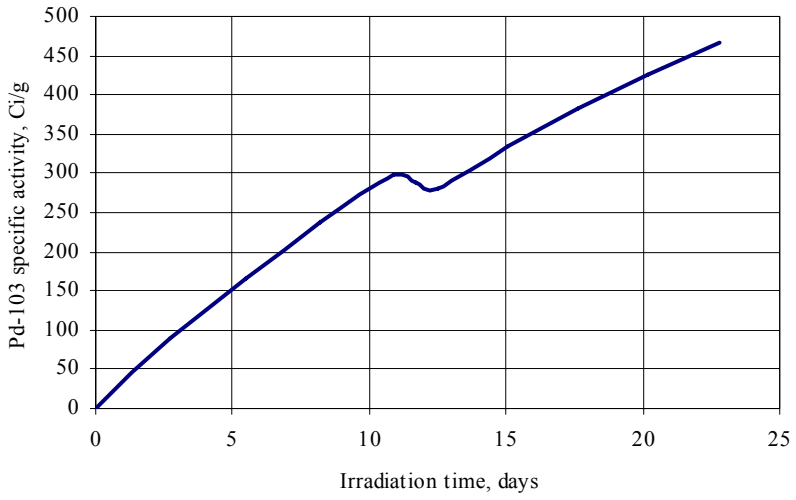

FIG. 4. Irradiation time dependence of ^{103}Pd specific activity

In our experiments the measured activity of palladium-103 was 980 mCi (36,3 GBq) at the end of irradiation; that corresponds to palladium-103 specific activity of 495 Ci/g. Mass-spectrometry measurements were applied to the irradiated sample to check the gamma-spectrometry value. The results of this experiment proved that the content of the palladium-103 was less than 0,5%, which corresponds to the specific activity of 640±190 Ci/g.

This result is in a good agreement both with the activity measurements and calculations. Figure 4 presents the calculated dependence of palladium-103 specific activity on the irradiation time in the neutron trap of the SM reactor for actual reactor operation mode.

The target was processed for 5 days after irradiation, and gamma-spectrometry was carried out on the 6th day. No measurable (at the level exceeding 0.01%) activity of palladium-101 and silver-111 radionuclides was detected. Probably, this is due to a low yield of these radionuclides after irradiation of highly enriched palladium-102. Besides, these radionuclides (even their formation is most probable) have short half-lives and they could completely (i.e. below the detection limit) decay by the moment of the activity counting. So, a possibility was demonstrated to produce palladium-103 with specific activity of ~500 Ci/g (~ 18.5x10^3 GBq) by irradiation of a highly enriched target in the high-flux research reactor SM.

4.2. Selection of an electrolyte composition for palladium plating and cathode pre-treatment

A procedure for electrochemical plating of palladium-103 onto a metal support was worked out under this Project that could be applied for the development of the technology of palladium-103 sources production. The development of the electrochemical procedure should also include selection of the electrolyte composition and the support material. The criteria used for the selection were the current palladium yield and the quality of the plated layer. Selection of electrolyte, in its turn, was based on the following criteria:

- simplicity and ease of preparation;
- stability of the electrolyte;
- minimal losses of palladium-103;
- possibility to correct the composition;
- production of uniform and good adhesion of palladium layers.

4.2.1. Aminochloride electrolyte

This electrolyte is frequently used for palladium plating in industry and research. Taking into account that handling of highly concentrated palladium-103 solutions (recommended concentration is 35 g/L) is not reasonable, we have focused our study on the possibility to work with a lower palladium initial concentration (C_{Pd}^{o}). Our results demonstrated that palladium can be successfully plated at the concentration C_{Pd}^{o} decreased down to 0.5 mg/ml. At C_{Pd}^{o} = 0.4 mg/ml the layer is dark-colored and spotty (non-homogeneous). The decreased cathode current could possibly further decrease palladium concentration in the electrolyte. However, a number of principal disadvantages of aminochloride electrolyte were highlighted during our research:

- solution pH drops down during the electrolysis; the pH value is required to be within the range of 8-10, decreased pH to 7 causes precipitate formation;
- preparation of the electrolyte is accompanied by formation of some precipitate that should be filtered off and causes losses of palladium-103;
- complicated procedure for electrolyte preparation (via Vockelen salt formation) excludes possibility to correct composition of the solution that also gives losses of palladium-103.

4.2.2. Nitrite electrolyte

Palladium precipitation on the cathode is very fast, but its layer has weak adhesion to the support. The decrease of initial concentration of palladium improved the quality of the layer, but attempts to produce completely non-leachable layer were failed.

4.2.3. Phosphate type electrolyte

Precipitation of palladium from this electrolyte gives uniform layers with a good adhesion to the support. The phosphate type electrolyte has no disadvantages inherent to the aminochloride electrolyte, the layer quality is good enough, and so this electrolyte is used in further experiments. Results of the study of palladium distribution are presented below in table I and Figs.5-13.

4.2.4. Pre-treatment of cathode before palladium plating

A selection was made out of palladium and nickel wires of 0.3 mm diameter. These materials were selected because together with palladium they are in the same group of Periodic System and have a face-centered cubic lattice similar to the electroplated isotope. It should provide a good adhesion of the palladium layer to the support.

TABLE I. PLATING CONDITIONS AND CATHODE PARAMETERS

Exp. No.	Cathode No.	$A^{103}_{Pd//}/C_{Pd}$ In electrolyte, mCi/mg	Plating time, min	Length of active cathode (core), mm	$A^{103}_{Pd\ (cathode),}$ mCi
1	1	0,91	30	31	0,52
2	2	0,87	90	28	1,09
3*	3-1	1,02	150	32	1,76
	3-2			32	1,61
	3-3			32	1,63
	3-4			32	1,56
	3-5			32	1,59
4	4	0,91	70	27	0,77
5	5	1,03	70	30	0,72
6	6	1,13	70	30	0,34
7*	7-1	2,54	60	29	1,13
	7-2			29	1,27
	7-3			29	1,24
	7-4			29	1,24
	7-5			29	1,22
8*	8-1	2,28	120	30	2,53
	8-2			30	2,18
	8-3			30	2,25
9	9	2,25	60	30	0,59
10	10	2,25	120	28	1,00
11	11	2,25	120	31	1,02

* cell N 2 was used in the experiment

Before palladium electroplating the support surface was treated (activated) with electrolyte of the following composition:

- 20 ml H_2O
- 2 ml HNO_3 (conc.)
- 2 ml HCl (conc.).

It was experimentally found that the nickel wire was effectively activated for 30 sec at 50^{o}C or 1-2 min at ambient temperature. So, the nickel wire was treated at room temperature during 1-3 min in further experiments. Palladiums wires were etched with ethanol and treated with similar electrolyte for 5 min. Supports were flushed with water and ethanol after chemical treatment.

4.3. Palladium distribution along the cathode

After plated with palladium-103, the cathodes were scanned using a domestically made device equipped with a collimator (the collimator's window was 1 mm wide) to examine the uniformity of the plated palladium. Scanning was performed both in longitudinal and radial directions of the cathode surface. Results for some cathodes are presented in Figs.5-12. The measured value was referred to the average one for the cathode, in percent. The point of coordinate origin corresponds to the bottom of the cathode.

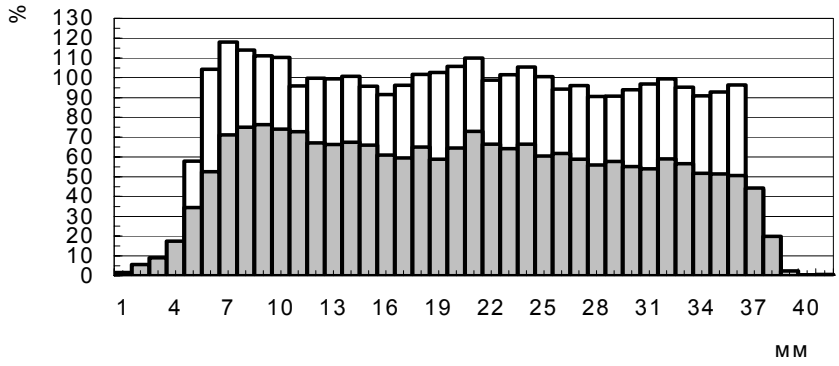

FIG. 5. Cathode N1 scanning, with and without titanium shield

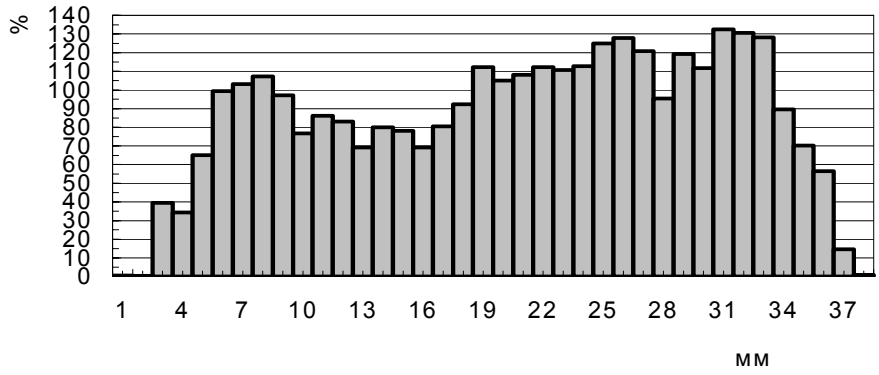

FIG. 6. Cathode N 3-2 scanning

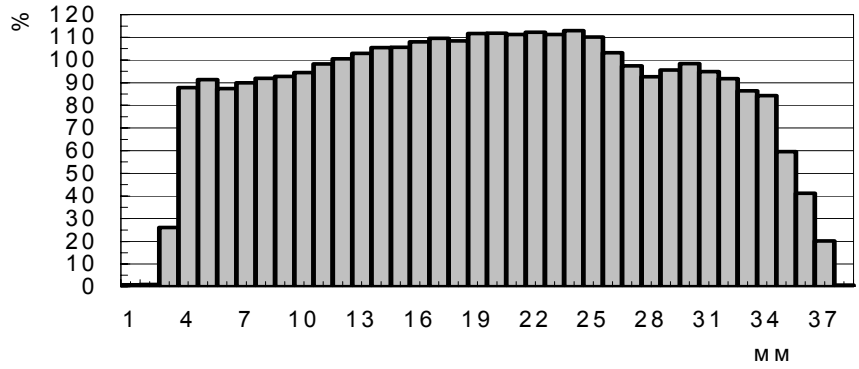

FIG. 7. Cathode N 3-3 scanning

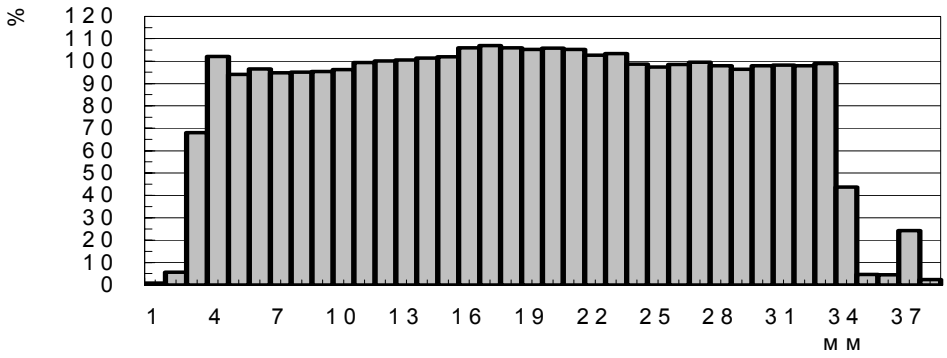

FIG. 8. Cathode N 8-1 scanning

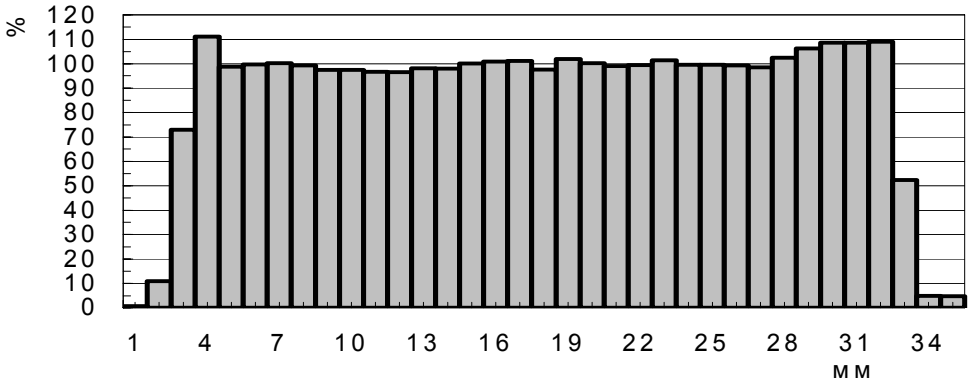

FIG. 9. Cathode N8-2 scanning

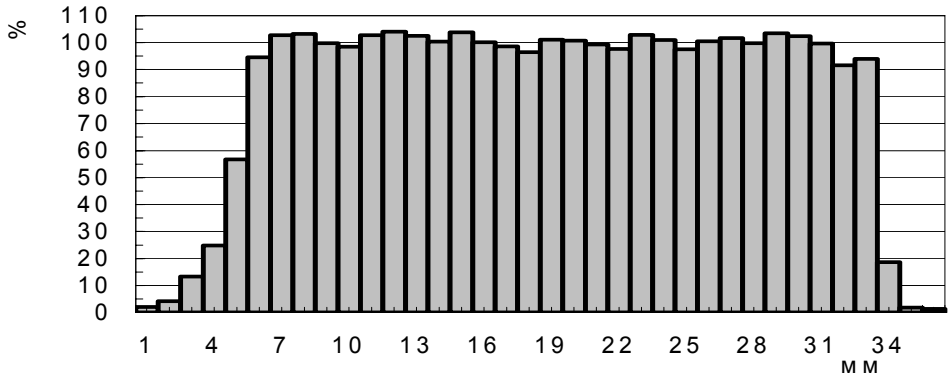

FIG. 10. Cathode N 10 scanning

As can be seen from the data presented in Figs.5-10 distribution of palladium-103, both in longitudinal and radial directions, is not high. It depends on a number of factors, like:

- formation of gaseous products during electrolysis and adhesion of bubbles to the cathode surface,
- non-uniform passivity of the cathode surface.

Gaseous bubbles adhesion to the cathode surface can not be eliminated even in cell 2 with forced electrolyte mixing. It was found that the quality of the cathode surface has more effect on the uniformity of palladium distribution. Chemical pre-treatment of the cathode, even a modified one (increased etching time, using of more effective etching solution), does not guarantee the uniformity. More effective is a combination of sintering in vacuum before chemical treatment. Figures 11-13 demonstrate a good radial distribution of the palladium plated in the cell 2 with flat anodes.

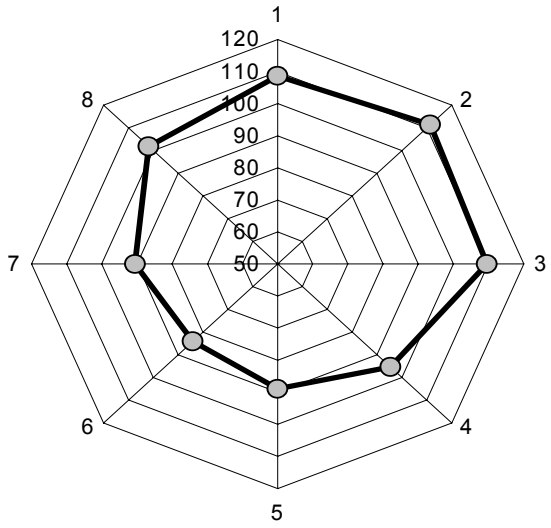

FIG. 11. Radial distribution of ^{103}Pd activity, cathode N 2;measured at 45^{o} rotation of the cathode

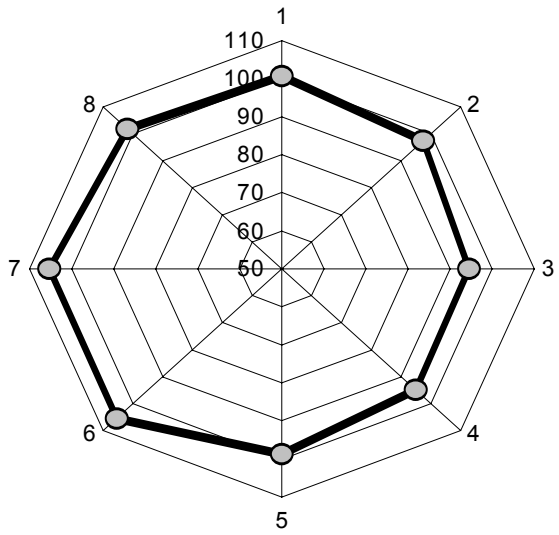

FIG. 12. Radial distribution of ^{103}Pd activity, cathode N 8-2;measured at 45^{o} rotation of the cathode

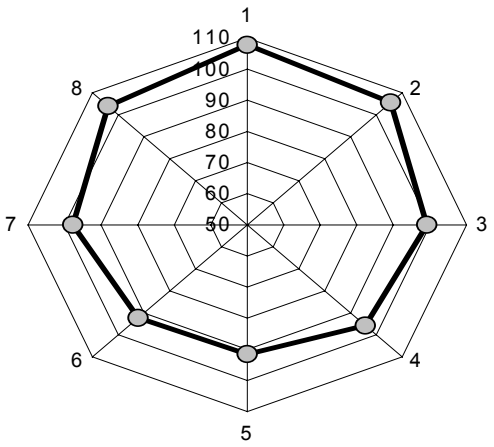

FIG. 13. Radial distribution of ^{103}Pd activity, cathode N 11;measured at 45^{o} rotation of the cathode

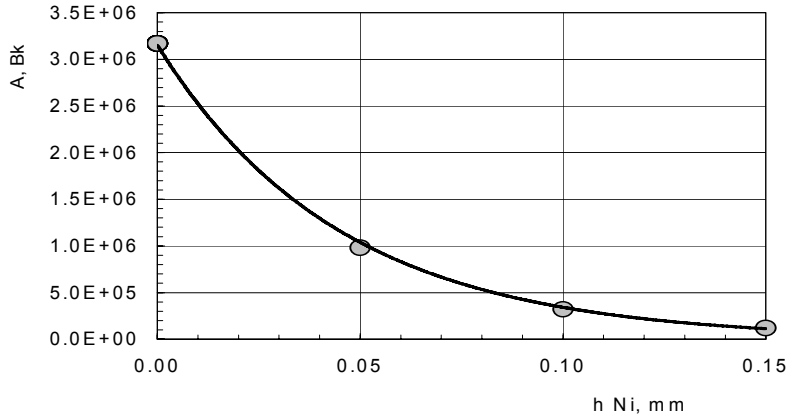

FIG. 14. Dependence of absorption of ^{103}Pd radiation on the thickness of nickel

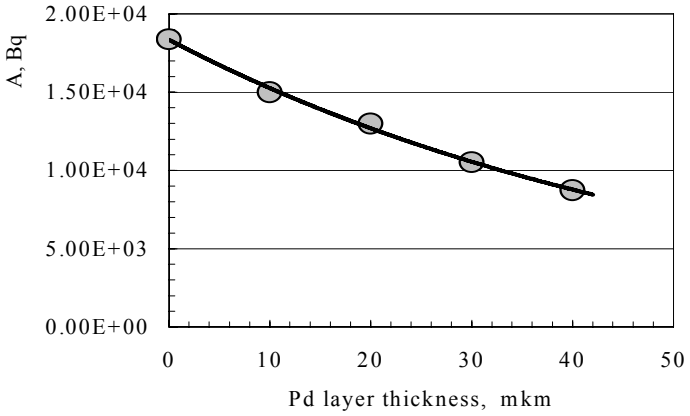

FIG. 15. Dependence of absorption of ^{103}Pd radiation on the thickness of palladium

4.4. Evaluation of calibration factors for palladium-103 sources activity measurements

4.4.1. Absorption of palladium radiation by the wall of the source titanium capsule

Absorption of palladium radiation by the wall of the source titanium capsule was evaluated as follows. The activity of palladium-103 plated on the cathode placed inside the titanium tube of 0.8 mm diameter with the wall thickness 0.05 mm was compared with that one measured directly. Besides, mock-ups of sources were prepared by cutting the cathode into pieces 3 mm long; they were placed inside the titanium capsule, with one end pre-welded, and sealed by a stopper inserted into another end. Activity of 3 mm cathode was measured before and after sealing. It was found that capsule wall absorbs 34±6% of palladium radiation.

4.4.2. Absorption of palladium radiation in the cathode

A portion of palladium-103 radiation is absorbed by the cathode – the source core. To evaluate absorption correction factor we measured dependency of ^{103}Pd activity on the thickness of the absorbing layer of nickel (the cathode material) as well as self-absorption in palladium (Figs.14,15).

From dependency of radiation absorption by nickel a correction factor was found to be (calculated) $K = A_{full}/A_{measured} = 1,89$. To check this value palladium was leached out of the cathode and its activity was measured. The correction factor was found to be equal 1.75 that is close to the calculated value. Notice, that in case of using a cyclotron producing carrier-free palladium-103 (specific activity of at least 6000 Ci/g) the palladium layer is at least 1.5 micrometers. In this case, the self-absorption factor will be less than 3%.

5. CONCLUSIONS

- A possibility to produce palladium-103 of 495 Ci/g (1.8×10^4 GBq/g) specific activity as of irradiation completion by irradiation of isotopically-enriched palladium-102 in the high-flux research reactor SM is demonstrated.
- A technique for palladium-103 electrochemical deposition from phosphate electrolytes on a nickel wire is proposed.
- A thorough preparation of the cathode surface is required for proper deposition of palladium.
- Correction factors for calculation of the activity of the produced sources with respect to palladium-103 self-absorption in the cathode material and the source protective capsule are determined.

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