

TECHNICAL REPORTS SERIES NO. 432

**Standardized High Current
Solid Targets for Cyclotron
Production of Diagnostic
and Therapeutic
Radionuclides**



IAEA

International Atomic Energy Agency

STANDARDIZED HIGH CURRENT
SOLID TARGETS FOR CYCLOTRON
PRODUCTION OF DIAGNOSTIC
AND THERAPEUTIC
RADIONUCLIDES

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INTERNATIONAL ATOMIC ENERGY AGENCY
VIENNA, 2004

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FOREWORD

Radionuclides continue to play an important role in diagnostic and therapeutic applications in modern nuclear medicine. Many of these radionuclides are produced in cyclotrons and the corresponding laboratory production methodologies and technologies are subject to constant improvements. In particular, ^{201}Tl , ^{123}I , ^{124}I and ^{103}Pd are important radioisotopes for use in medical diagnosis and therapy and it is expected that their use will increase in the foreseeable future. Thallium-201 is already being regularly produced and utilized in several countries, including a number of developing countries. Iodine-123 is widely used for imaging by single photon emission tomography where $^{99\text{m}}\text{Tc}$ analogues are not yet available, for example for brain receptor imaging, whereas ^{103}Pd has recently been successfully introduced for brachytherapy of prostate cancer and other solid tumours. Iodine-124 is a potentially useful radionuclide for targeted therapy as well as for molecular imaging by positron imaging tomography. Interest in the latter two isotopes is growing amongst many developing countries operating cyclotrons.

The IAEA has been supporting, through its various programmes, activities related to increasing cyclotron production of radioisotopes. On the recommendation of several experts, a Coordinated Research Project (CRP) on Standardized High Current Solid Targets for Cyclotron Production of Diagnostic and Therapeutic Radionuclides was begun in 2000 with a duration of three years. The aim of the CRP was the optimization and standardization of solid phase cyclotron target technology for the production of ^{123}I , ^{124}I , ^{201}Tl and ^{103}Pd with a view to increasing production yields, improving the economics of production and the availability of these four important radionuclides. Scientists from Argentina, Belgium, China, Hungary, Indonesia, the Islamic Republic of Iran, Kazakhstan, Romania, the Russian Federation, Saudi Arabia and the United States of America participated under research agreements and contracts. The laboratory protocols developed under the auspices of the CRP and included in this report are considered effective as well as simple to follow by anyone wishing to embark on the production of radionuclides using solid target technology, not only for the above mentioned four radionuclides, but for other cyclotron produced radionuclides as well.

The IAEA wishes to thank all the participants in the CRP, as well as their collaborators, for their valuable work and scientific contributions. Thanks are due to P. Van den Winkel for sharing his expertise, as well as for his contribution to the preparation of this report. The IAEA officers responsible for the CRP and this report were H. Vera Ruiz, D.V.S. Narasimhan and M. Haji-Saeid of the Division of Physical and Chemical Sciences.

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1. INTRODUCTION

Radioisotopes produced by cyclotrons, and their corresponding radiopharmaceuticals, have been shown to be extremely valuable in basic medical research, disease diagnosis and radiotherapy treatment. More than 300 cyclotron facilities exist worldwide and the number is growing. A number of IAEA Member States have acquired cyclotrons to produce radioisotopes for nuclear medicine and several others have expressed an interest in acquiring such facilities.

Many of these facilities support strong and active radionuclide production programmes to meet the ever increasing demand for diagnostic and therapeutic radionuclides from modern nuclear medicine. In particular, the number of cyclotron facilities starting to produce such recently introduced radionuclides as ^{124}I for possible dual use in diagnostic and therapeutic procedures, and ^{103}Pd for brachytherapy of prostate cancer and other solid tumours, is dramatically increasing as a recent survey conducted by the IAEA revealed [1]. Additionally, the demand for two other more classical radionuclides, ^{123}I and ^{201}Tl , also continues to rise within the nuclear medicine community.

Concurrent with this, or as a consequence, the need to optimize further cyclotron procedures and target technologies has become evident. Of particular significance is the fact that the above mentioned radionuclides can be produced in available cyclotrons in Member States using solid target technology.

The Coordinated Research Project (CRP) on Standardized High Current Solid Targets for Cyclotron Production of Diagnostic and Therapeutic Radionuclides focused on the optimization and standardization of solid phase cyclotron target technology for the production of ^{123}I , ^{124}I , ^{201}Tl and ^{103}Pd . In particular, the main technical goals of the CRP, as set up at its inception, are the following:

- (a) To investigate the possibility of using electrodeposited tellurium and melted tellurium oxide as the target material for the production of ^{123}I and ^{124}I . For the oxide target, the following parameters and techniques will be explored:
 - (1) Methods of deposition of the oxide on the metal target plate;
 - (2) The stability of the oxide layer during storage and irradiation;
 - (3) The thermal characteristics of the oxide layer, especially during irradiation;
 - (4) Methods of radioiodine recovery such as dry distillation;
 - (5) Reprocessing of the targets.

- (b) To prepare a target for the production of ^{201}Tl which will withstand high beam currents in the range of 100–300 μA . This will be accomplished through transfer of the technology available on the electrodepositing of thallium onto copper plates to produce a smooth, stable surface. The techniques will include preparation of the targets before irradiation, extraction of the desired radioisotope, and processing and recovery of the enriched materials after irradiation. Furthermore, a standard procedure for the preparation of trivalent ^{201}Tl ($^{201}\text{Tl}^{3+}$) suitable for polypeptide labeling has been developed.
- (c) To prepare a target for the production of ^{103}Pd from rhodium. This will be accomplished by electrodepositing Rh onto metal plates. The chemical removal of Rh was also investigated as an integral part of the CRP. Although not an essential goal of the CRP, attempts were made to investigate the production and quality control methods for the preparation of sealed sources for treatment of prostate cancer.

It was anticipated that the results of the CRP would contribute to strengthening existing radionuclide production programmes in Member States operating cyclotrons and increasing the availability of high quality radionuclides through the use of solid target technology.

Experienced scientists from developing and developed countries participated in the CRP from 2000 to 2003. Scientists from Argentina, Belgium, China, Hungary, Indonesia, the Islamic Republic of Iran, Kazakhstan, Romania, the Russian Federation, Saudi Arabia and the United States of America participated under research agreements and contracts.

The report resulting from the first Research Coordination Meeting (RCM) includes details of the research work plan agreed upon for the first 15–18 months and also includes recommended outlines of laboratory protocols for the participants to follow and improve during this period. In particular, it was agreed to further investigate the following parameters:

- Beam diagnostic and beam quality;
- Target preparation;
- Target cooling;
- Irradiation geometries;
- Nuclear reactions and nuclear data;
- Required isotopic composition;
- Target backing materials;
- Geometry of the target;
- Plating of the targets;
- Melting;

- Target processing and recovery;
- Quality control of the fabricated targets;
- The final radionuclide product.

The second RCM brought substantial progress in all the above-mentioned parameters, which warranted continuation of the CRP. Co-operation in joint research among the participants was also developed. In addition, several areas were identified for further investigation, including the thermal diffusion of ^{103}Pd from rhodium foils, the identification and quantification of impurities in the proton irradiated rhodium, the magnitude of the contribution from the $^{123}\text{Te} (p \gamma) ^{124}\text{I}$ reaction and the development of quality control procedures for the electroplated targets.

At the third and final RCM, final reports of the research contract and research agreement holders were presented. The scientific results reported indicate that the original goals of the CRP were fully achieved. Improved standardized methodologies and procedures for high beam current solid targets for cyclotron production of ^{123}I , ^{124}I , ^{103}Pd and ^{201}Tl were developed and tested, including quality control parameters, improved target beam irradiation, target fabrication and target processing, and recovery strategies. This optimization ultimately leads to increased high quality radionuclide production yields for medical use, at lower cost. The individual, unedited reports of the participants as they were presented at the third RCM are included with this book on a CD-ROM.

2. SUMMARY OF THE ACHIEVEMENTS OF THE CRP

In general it was found that, apart from the melting point of the target metal, the critical factors determining the maximum allowable beam current which may be applied to the targets are the cooling of the target carrier and the beam current density distribution on the target. The thinner the copper plate and the higher the water flow velocity that can be achieved (high Reynolds number), the higher the maximum beam current burden that can be tolerated. For optimum target irradiation, this should be accompanied by a reduction of the maximum beam current density that can be achieved by linear beam scanning or circular wobbling of the beam. An appropriate beam diagnostic device should then be available. The major technical and scientific developments achieved by the participants are summarized in the following sections:

2.1. THALLIUM-201

- (a) Plating from environmentally friendly (cyanide free) solutions has proven to give excellent Tl target layers if the composition and metal contents of the plating bath, the plating waveform and method, the plating vessel geometry and the plating current density are adjusted properly.
- (b) When the plating current density is set to 2–3 mA/cm² and the ²⁰³Tl concentration is adjusted so that at the end of the plating the metal concentration is not lower than 16.2 g/L ‘critical concentration’ (C_c), time controlled production (requiring 100% plating current efficiency (CE)) of as many as 8 targets in less than 6 h is feasible. As the plating method, AC bipolar chopped sawtooth waveform (with $f_{ST} = 100$ Hz, $f_{CH} = 1000$ Hz) constant current electrolysis (CCE) from alkaline EDTA baths is applied using multi-window cylindrical plating vessel geometry with a single, axial and grounded platinum wire anode.
- (c) A linear relationship between the maximum allowable plating current density and the critical ²⁰³Tl concentration was established. It allows calculation of the input concentration of ²⁰³Tl in the plating solution and of the plating time required for time controlled production of the desired layer thickness. A programme allowing calculation of the optimum plating conditions was being developed.
- (d) An improved water coolant channel geometry (flat copper carrier, no cooling fins, narrow coolant channel 2.5 mm wide with a rectangular cross-section area of 0.3 cm²), through which a coolant flow rate with a velocity higher than 2600 cm/s is forced, has been designed to allow the target to be operated at higher (theoretically as much as 320 μA) beam currents.
- (e) A program based on the calculated maximum allowable target beam current density has been developed that allows calculation of the optimum wobblers’ radius when circular wobbling is applied.
- (f) Efficiently cooled ²⁰³Tl layers (thickness 80 μm) have been demonstrated to withstand at least 270 μA for 9 h with no apparent loss of material when irradiated in a 6° beam/target geometry with a circular wobbled beam. The maximum beam current density on target then amounts to 0.5 μA/mm².
- (g) A new recovery technology has been developed for the fast and easy separation of target material from processed target solutions. It involves controlled cathode potential electrolysis (CCPE) and allows the quantitative separation of Cu, ⁶⁵Zn and ²⁰³Tl (as much as 16 g in a single run) within six hours. Copper and zinc are electrodeposited on a large area copper gauze while thallium is collected on a platinum gauze

electrode, from which a new ^{203}Tl plating bath can be prepared simply. The reported plating/recovery/cooling and irradiation mode technology is the result of close collaboration between Belgium, the Islamic Republic of Iran and Kazakhstan.

- (h) A new procedure has been implemented in Argentina that utilizes the information generated by the CRP participants. This has resulted in a high purity product and completely automated operation. An ion exchange chromatography method for the separation of $^{201}\text{Tl}^+$ from Pb^{2+} has been developed and implemented.
- (i) The transfer of technology from the CRP made it possible for Kazakhstan to start producing and distributing ^{201}Tl as a new radionuclide product. The ^{203}Tl recovery rate in Kazakhstan has been improved to 99.5%.
- (j) A program has been developed in Hungary which allows prediction of the levels of radionuclidic impurity, based on the isotopic composition of the target material and using the best values for the nuclear reaction cross-sections for the relevant reactions.
- (k) Quality control tests have been developed for electroplated ^{203}Tl targets. They include a chemical metal purity (CMP) test, surface area granulometry (SAG) test, layer homogeneity test and thermal shock test (TST). Apart from the latter, all quality control tests can be performed on-line. The TST should be considered as a quality control test on the plating technology and should be tried with natural target material.

2.2. IDODINE-123 AND IODINE-124

- (a) Based on the information generated in the CRP, a remote controlled system has been designed for the production of ^{123}I from the TeO_2 target. This system has been installed in the Russian Federation.
- (b) To improve heat removal and enhance the maximum beam current burden of TeO_2 targets, a very efficient helium cooling system on the target beam side and a beam scanning device were introduced. It allows a maximum beam density current of 6–8 μA for a 250 mg/cm^2 TeO_2 layer irradiated for at least 14 hours in a 90° beam angle geometry.
- (c) Quality control tests have been developed for the TeO_2 targets. Upon preparation, the target layer should be checked visually for colour and uniformity of the layer thickness. This inspection should be repeated and the radionuclidic yield and purity estimated after a short (one hour) irradiation before the long term (>12 h) production irradiation.
- (d) The technology for plating tellurium metal on nickel coated copper target supports from alkaline ($\text{pH} > 10$) plating baths, containing no quality

improving additives and prepared from TeO_2 , has been optimized in China and Saudi Arabia. Application of DC CCE at 10 mA/cm^2 for 1 h in appropriate plating geometry results in $20 \mu\text{m}$ layers showing excellent homogeneity and surface granulometry. Bombarded with 25 MeV protons a mean, non-wobbled beam current density of as much as $8 \mu\text{A/cm}^2$ can be tolerated with limited losses ($<3\%$) of enriched material.

- (e) Optimization of the recovery (precipitation/distillation) procedure in China resulted in a 99% increase in recovery of the expensive enriched tellurium.
- (f) By changing the thickness of the target carrier plate to 0.5 mm as recommended by CRP participants, the maximum beam current burden was increased from $30 \mu\text{A}$ to $80 \mu\text{A}$ ($8 \mu\text{A/cm}^2$) in China.
- (g) A program has been developed in Hungary which allows prediction of the levels of impurity based on the isotopic composition of the target material and using the best values for the nuclear reaction cross-sections for the relevant reactions.

2.3. PALLADIUM-103

A new plating/electrodissolution/recovery cycle was developed in a collaborative effort of Belgium and the Islamic Republic of Iran.

- (a) Rhodium targets produced from home prepared rhodium sulphate solutions ($\text{pH} = 1-2$) prepared from recovered hydrated Rh_2O_3 and containing 1% sulphamic acid as stress reducing agent show a quality (homogeneity, surface granulometry, TST) comparable to targets obtained with commercially available and expensive baths when DC CCE plating (10 mA/cm^2) to depletion and at $40-60^\circ\text{C}$ is performed.
- (b) A new centrifugal AC electrodisolution system was developed. It allows quantitative ($>99\%$) and time controlled solubilization at 80°C of as much as 3 g of irradiated rhodium fragments, cut wires, foils and powder within 3 h in a limited volume (40 mL) of concentrated hydrochloric acid. The system operates at 2 A/cm^2 and is fitted with appropriate feedback devices for volume, temperature and current control.
- (c) The best chemistry for the recovery of rhodium was evaluated and results indicated that hydrated Rh_2O_3 precipitation works best.
- (d) The thermal diffusion technique (Argentina, Kazakhstan) for the extraction of ^{103}Pd from irradiated rhodium foils has been further evaluated. As such a process would allow minimizing the target

preparation time, simple ^{103}Pd extraction chemistry and reuse of the target foil, further research is worthwhile. At present extraction yields after heating at 1400°C for 30 min in hydrochloric acid are limited to a maximum of 70%.

- (e) Methods of preparing brachytherapy seeds on a small scale have been tried in both Argentina and China.
- (f) Argentina has directly irradiated rhodium wire with the intention of producing seeds directly from the wire. The problems encountered were production of radionuclidic impurities of rhodium and the self-shielding of the emitted radiation by the rhodium wire.
- (g) A dedicated beam line has been installed in Romania for the production of ^{103}Pd from rhodium. Further development is under way for the production of seeds.

Additionally, a target quality test procedure has been developed for all the targets. Essentially, four tests should be performed.

The first is a chemical purity test on the plating solution and/or on the recovered target material before plating. Multielement analytical techniques should show a chemical purity of more than 99.5% as far as the target metal is concerned.

The second test is a thermal stability (thermal shock) test in which the target plate is heated for 1 hour to 50° below the melting point of the metal with the lowest melting point (target or carrier metal) and then plunged into a cold-water bath. No crater formation or peeling off of the target layer should be observed.

The third criterion deals with a SAG test. The result of a microscopic examination (microscopy, scanning electron microscopy (SEM)) of the target layer surface should be compared with that of an accepted standard reference. 'Comparable with' should be used as the criterion for an allowable grain size distribution.

The last test is evaluation of the homogeneity of the layer thickness. This may be done using an ultrasound device or by micrometry if the more sophisticated technique is not available. A standard deviation (SD) of 5% on the mean thickness is acceptable.

3. OUTPUTS AND OUTCOMES OF THE CRP

The basic and fundamental aspects of the electroplating technology for the preparation of cyclotron solid targets were imparted to all the participating scientists in the initial phase of the CRP. In addition, the participants have improved or developed new methods of producing, purifying and recovering the target materials. During the course of the CRP, several new topics were identified for investigation and the results of these investigations are included in this report.

3.1. RESULTS

Several results of the CRP are worth mentioning. Some of these apply to all radionuclides investigated under the auspices of the CRP (^{123}I , ^{124}I , ^{103}Pd and ^{201}Tl), others are more specific to each radionuclide.

Most laboratories have applied the plating technology developed by P. Van den Winkel to their own targets and have correspondingly improved production yields and reliability, thus contributing to the efficiency and relevance of their radioisotope production and distribution efforts. The overall efficiency of the operational steps has likewise increased, e.g. the number of targets that can be simultaneously plated can be increased and the number of rejected target plates has been considerably decreased. In addition, a program, PLATING.EXE, has been developed and put at the disposal of the participants. It allows fast calculation of the ^{203}Tl plating parameters (input concentration, plating current and time) for time controlled multi-target plating. The effect of heat transfer out of the target has been evaluated in several places and new target carrier cooling geometries were designed and fabricated accordingly. Moreover a code, OPTIWOB.EXE, that generates the optimum conditions for circular beam wobbling, was developed in Belgium. These new designs and procedures have allowed the beam current to be dramatically increased. This may be put on the target, resulting in a better production yield.

Microsoft Excel based software to estimate radionuclide purity as a function of target isotopic purity was passed on by Hungary. This program allows calculation, before irradiation, of the radionuclidic purity of the final product as a function of time at the end of bombardment. A program that calculates stopping power and radioisotope yields was given to the participating countries by the USA.

3.1.1. Thallium-201

An automated system for the recovery of thallium has been developed and installed. The recovery of thallium has been significantly improved with the use of electrochemical techniques. These techniques have been applied in some of the participating laboratories, with the result that more of the expensive enriched target material can be recovered. As a result the supply of enriched target material does not have to be replenished as often as previously.

Software and interface electronics for computer generation of plating voltage waveforms for solid target preparation were developed and distributed to the participants. Plating performed from cyanide free solutions has proven to give an excellent product with proper adjustment of the metal contents of the plating bath. The maximum allowable power dissipation on plated targets has been increased significantly. A beam current of 270 μA for 9 h has been applied to a thallium target with no apparent loss of material. A very efficient but simple ion exchange chromatography method for the one step separation of $^{201}\text{Tl}^+$ from $^{201}\text{Pb}^{2+}$ has been developed, automated and implemented under this CRP.

3.1.2. Palladium-103

Under this CRP, a method for the thermal diffusion separation of ^{103}Pd from a ^{103}Rh foil was developed which would allow the non-destructive recovery of ^{103}Pd from the foil. This technique, if perfected, could allow the reuse of a single metallic foil for multi-batch production of the radionuclide. The nuclear cross-section for the production of ^{103}Pd from the $^{103}\text{Rh}(d,2n)^{103}\text{Pd}$ nuclear reaction was measured and the feasibility of producing ^{103}Pd from this reaction was assessed. In a collaborative effort between Belgium and the Islamic Republic of Iran, a cylindrical graphite centrifugal electro-dissolution mini-reactor was developed which allows effective quantitative and time controlled solubilization of small pieces of irradiated rhodium. This was a new breakthrough in the technology for the plating of rhodium targets and recovery of the rhodium metal, i.e. in the plating/irradiation/dissolution/recovery cycle.

The wet chemistry for rhodium recovery was evaluated and it was determined that Rh_2O_3 works best. Two countries, as a result of their participation in the CRP, managed to produce ^{103}Pd seeds for brachytherapy applications. The CRP thus demonstrated that this technology is well within the capabilities of developing countries and that brachytherapy seeds can be produced on a small scale and made available for clinical applications.

3.1.3. Iodine-123

A reusable tellurium oxide target has been used to produce ^{123}I . The ^{123}I is removed from the glassy matrix with dry distillation heating. The factors affecting the purity of the final material and the stability of the target have been evaluated and a series of quality control tests have been developed. Based on the ^{124}I radionuclidic impurity in the ^{123}I obtained from very highly enriched ^{123}Te , the $^{123}\text{Te}(p,\gamma)^{124}\text{I}$ reaction has been postulated as the source of this impurity. These preliminary results indicate a limit to the purity of the ^{123}I produced by this method.

It has also been shown that the key to the level of impurity of ^{124}I from the $^{123}\text{Te}(p,n)^{123}\text{I}$ reaction is the amount of ^{124}Te in the initial material. A remote controlled system for the production of ^{123}I from TeO_2 has been built as part of this CRP. A beam scanner has been used to evaluate the effect of beam wobbling and beam optics on yield. The technology for plating of tellurium on nickel coated copper has been used and loss of tellurium in the beam was reduced with this technology. A new method of preparing ^{123}I from a tellurium metal target, utilizing the technology developed in this CRP, has been used. The new method has increased yields by more than 30% with the same quality final product.

3.2. OUTCOMES

Improved cyclotron solid target technology has been developed under the auspices of the CRP. This has allowed an increase in both the quality and reliability of the production of ^{103}Pd , ^{123}I and ^{201}Tl in the participating laboratories. Having at hand improved and more reliable technologies has increased the practical radionuclide production yields by as much as 30% in some instances, contributing to the efficiency and economy of radionuclide production programmes.

4. OPTIMIZATION OF THE PLATING TECHNOLOGY, COOLING AND IRRADIATION MODE OF ELECTROPLATED SOLID TARGETS, AND QUALITY CONTROL OF SOLID TARGETS

4.1. INTRODUCTION

To be up to standard, electroplated solid targets should meet a series of physicochemical quality requirements and should satisfy a set of demands concerning the implementation of plating technology for the industrial production of radionuclides. However, when they are irradiated with particle beams showing a poor beam current density distribution, and/or an inappropriate cooling system is used, even high quality targets can give rise to losses of expensive target material during the bombardment, poor reproducibility and/or low yields. Therefore, preparation of high quality targets should be accompanied by an optimization of the target cooling and of the target irradiation modes.

4.2. QUALITY REQUIREMENTS AND OPTIMIZATION OF THE ELECTROPLATING TECHNOLOGY

4.2.1. Quality requirements and industrial demands

The major physicochemical quality criteria may be summarized as follows. First, the target layer should adhere strongly to the target carrier up to an irradiation temperature at least several tens of degrees below the melting point of the target metal. Secondly, it should be smooth (not spongy and dendrite free), dense (no occlusions or vacuoles), stress free and homogeneous ($\pm 5\%$), and show a well defined thickness that may vary from a few tens to several hundred microns, depending on the shape of the excitation curve and on the beam/target angle geometry. Finally, the metal deposit and the carrier/layer interface should be free of traces of organic plating bath additives such as complexing reagents, surfactants or stress reducing agents. Poor quality of target layers means that one or more of these conditions has not been satisfied.

The main causes of poor physicochemical quality are:

- (a) Gas (hydrogen) evolution at the cathode during plating, resulting in spongy and irregular deposits;

- (b) Concentration polarization of the electro-active species (i.e. the hydrated or complexed target metal ion), giving rise to treelike structures called dendrites and requiring post-plating mechanical fashioning as pressing or rolling, resulting in an enhanced risk of plating bath occlusions;
- (c) Ill-considered choice of additives such as organic complexing agents and surfactants, causing peeling off of the target layer and/or target damage (crater formation) on irradiation;
- (d) Asymmetric stirring and maladjusted plating temperature, causing inhomogeneous layers and dendrites;
- (e) Non-homogeneous electrical fields at the target carrier/plating solution interface and maladjusted plating voltage waveforms, causing inhomogeneous layers and dendrites.

In the framework of routine industrial production the plating solution, the electromechanical and electronic set-ups and the target carrier should currently meet a number of important additional requirements such as:

- (1) The plating solution must be free of toxic anions (especially CN^-) and well suited for repetitive plating (as many as ten batches per cycle) by simple replenishment of the metal contents of the bath after each batch. At the end of a plating cycle any enriched material remaining in a partially depleted bath must be recovered quantitatively in a physicochemical form that can be reused for the preparation of a new plating solution;
- (2) Both the plating bath and the set-up must allow multi-target preparation (minimum four per batch), preferably at room temperature, and with a duty cycle allowing at least one batch per shift (7 h) to be finished;
- (3) The thickness of the layer should be time controlled, meaning that the plating CE should be constant as a function of time, or better be 100%;
- (4) Post-plating mechanical fashioning (pressing, rolling) of the target layer must be excluded.

4.2.2. Optimization of the plating technology

From the above it follows that optimization of the plating technology for a given target metal involves the proper choice of plating method, the composition of the plating bath, the electromechanical set-up and temperature, and the plating voltage waveform and current density applied.

Plating methods

Constant voltage electrolysis (CVE), CCE and CCPE can be applied as plating methods.

Though requiring simple electronics and frequently applied in industry for thin layer film coating (as much as a few microns), the main disadvantage of CVE may be the variation of the plating current, and hence of the current density, as a function of time. As the current density affects the quality (crystal size, dendrite formation) of the deposit and an optimum current range density may be established for a given metal, thick layer (as much as a few hundred microns) deposition by this method will often result in poor quality. Moreover, as the CE is mostly less than 100%, time controlled plating may be a problem.

In CCE a fixed current is forced through the electrochemical cell and the applied plating voltage is automatically adjusted (i.e. increased) so that the set current is kept up. Using concentrated solutions of appropriate composition that show a limited depletion ratio (DR) for the electro-active species (as much as a few tens of per cent) at the end of the plating and applying an optimized plating current density, a 100% CE can be easily obtained. For time controlled layer thickness preparation, this technique is preferable.

Controlled cathode potential electrolysis involves the introduction of a reference electrode into the system. The potential of the cathode is set at a fixed value versus the potential of that reference electrode. This value corresponds to 99.99% depletion of the electro-active species, i.e. to the lower limit of its discharge interval.

The current decreases exponentially as the process progresses and its reading may be used to monitor the depletion of the bath. Controlled cathode potential electrolysis is well suited for quantitative separation of metals and is a very attractive tool in the recovery and purification of enriched target material.

Composition of plating solutions

In general, plating solutions contain a readily soluble inorganic salt (chloride, sulphate, perchlorate) and free acid (nitric, sulphuric, hydrochloric or perchloric acid) or base (sodium hydroxide, ammonia). In most cases, additives to improve the quality of the metal deposit are present. These compounds are complexing reagents, surfactants and cathodic or anodic depolarizers and stress reducing agents.

Hydrogen evolution at the cathode competing with the metal deposition may be reduced or even prevented by adjusting the pH of the solution. Whenever possible plating from strong alkaline medium should be applied, in

which case the proper choice of a complexing agent may result in a 100% CE, allowing time controlled plating.

Concentration polarization linked to plating current density and plating bath additives strongly affects the quality of the deposit. The higher the current density the smaller the crystal size and the better the physical properties of the metal layer. Smooth, strong and adherent deposits are made up of very fine crystals obtained at higher current densities. Moreover, higher current densities result in a higher over voltage for hydrogen, thus reducing the risk of gas evolution at the cathode. However, very high current densities do result in extremely high concentration polarization, leading to dendrite formation. From the foregoing, it follows that an optimum current density interval is to be expected for each metal. Its value may be increased and the plating time reduced by:

- (a) Plating from concentrated metal solutions so that the metal concentration at the end of the plating procedure remains high, meaning that the DR of the plating bath is relatively low. This strongly reduces or eliminates concentration polarization. From practical experience, it follows that a DR of as much as 40–50% is acceptable for many plating vessel and target window geometries. Appropriate replenishment of the partially depleted bath may then allow repetitive batch production with a single plating solution.
- (b) Proper choice of the valency and addition of non-incorporated complexing agents showing a low stability constant for the metal ion ($pK_{\text{stab}} =$ as much as 6–7) and for which the electron transfer rate at the solution/metal layer interface is high as compared with that of the hydrated M^{n+} cation. EDTA⁴⁻, formiate, acetate and citrate are examples of non-toxic organic complexing agents that can be tried. High stability constants will result in a large shift of the discharge interval of the metal towards more negative values with the possible consequence of hydrogen generation.
- (c) The addition of an excess of depolarizing agents. An anodic depolarizer such as hydrazine will fix the potential of the anode and avoids the evolution of oxygen. This prevents the oxidation of the metal ion to an unwanted higher valency and/or the re-oxidation of the plated out metal layer. The addition of a cathodic depolarizer such as nitrate will prevent the shift of the cathode potential and, in acid medium, prevent gas evolution.
- (d) The presence of a minor concentration of a tensio-active agent that causes a decrease of the surface tension of the solution and results in a decrease of the thickness of the hydrodynamic layer at the metal–solution

interface, causing enhancement of the transfer rate of the electro-active species (i.e. complexed or solvated metal ions) to the cathode surface and reduction of the concentration polarization. Though many organic surfactants (anionic, cationic or neutral) of acceptable purity are commercially available, the practical choice is very limited because most of them tend to stick at metal/target carrier interface or are incorporated in the metal lattice. Upon irradiation and heating up the organic material is carbonized, thus producing volatile decomposition compounds that lead to eruptions and crater formation, resulting in the loss of enriched material and a decrease in the irradiation yield. An example of a useful surfactant is BRIJ-35 (0.05% wt/vol.), a non-ionic polyoxyethylene ether, frequently used as a wetting agent in automated continuous flow analysers.

Electromechanical plating set-up and plating temperature

Simultaneous multi-target plating that results in homogeneous target metal layers requires an appropriate plating vessel design to ensure a homogeneous electric field all over the plating area and a well designed stirring system that avoids local concentration gradients and hence local concentration polarization of the electro-active species.

As far as the plating vessel is concerned, a cylindrical geometry is recommended. Indeed, the above requirements can be met by using a hollow Perspex cylinder with an appropriate diameter/height ratio (typically 1/3) and, for an even number of targets ($n = 4, 8$), a diameter/ $(n \cdot w)$ ratio of 1:25, where w equals the width of the plating area of a single target. The vessel should be fitted with a single, axial and grounded platinum anode wire (1 mm) mounted in the bottom and properly shielded at both extremities. Four (or eight) vertical slots in the outside wall of the cylinder allow the introduction of four target carriers. The geometry results in a quasi-homogeneous electrical field at the surface of the cathodes.

The stirring system should allow fast and efficient homogenization of the plating solution. This can be accomplished by introducing a hollow, perforated Perspex or POM cylinder surrounding the platinum anode and mounted on the axis of a DC motor-tacho combination with a variable rotation speed (up to 1200 rev./min) while the direction of rotation should be reversed periodically (typically every 8 s). The vigorous bi-directional stirring then reduces the thickness of the hydrodynamic layer and hence the concentration polarization. The geometry of the stirrer (length of the cylinder and number and diameter of the circular holes) needs some optimization for narrow ($d < 4.5$ cm) plating vessels.

The plating temperature may be an important quality parameter for some noble metals such as rhodium. At higher temperature the viscosity of the solution decreases and the mobility of the ions increases while in well stirred conditions, the thickness of the hydrodynamic layer is substantially reduced. Higher temperature hence reduces concentration polarization. On the other hand, over-voltages are lowered at higher temperatures, even for heavy metals such as thallium, the result being an increased risk of hydrogen gas evolution. Furthermore, the required electromechanical plating set-up should be much more sophisticated (heating element and associated feedback electronics for temperature control, solution level monitoring and feedback electronics to add water to compensate for evaporation). As is well known, the optimum temperature for a given metal electrolysis can only be determined experimentally. Whenever possible, plating at room temperature is preferable.

Plating voltage waveform and current density

In spite of an acceptable composition of the plating solution and an optimized electromechanical set-up for time controlled CCE, a slight tendency towards dendrite formation may persist when a DC waveform is applied. Often this phenomenon can be suppressed completely by the application of an AC voltage signal. During the cathodic part of the applied signal, deposition with micro-dendrite formation takes place. The metal is re-dissolved during the anodic part. As the anodic re-dissolution rate is higher for deposits showing a higher surface area to mass ratio, fine treelike dendrite structures will be solubilized faster. For a given metal, an appropriate choice of the waveform and its frequency (typically 100 Hz), of the amplitude of the cathodic and the anodic current (ratio typically 3/2) and of the duty cycle (fraction of time during which cathodic deposition takes place, typically 60%), may result in the ultimate elimination of dendrite formation. The optimum current density that should be applied strongly depends on the nature of the metal and its concentration and can vary between a few milliamperes per square centimetre (3 mA/cm² for Tl) up to several tens of milliamperes per square centimetre (40 mA/cm² for zinc).

Increasing the concentration of the metal and plating out to a lesser degree of depletion allows the plating current density to be increased. As an example, the practical conditions for the plating of four high quality targets are summarized in Table 1. In all cases, CCE is applied. With the exception of rhodium (40–60°C), all platings can be done at room temperature and involve the use of hydrazine as the anodic depolarizer.

TABLE 1. PLATING CONDITIONS FOR HIGH QUALITY TARGETS

Radionuclide produced	Plated target	Plating bath composition	Wave form applied	Current yield (%)
Tl-201	Tl-203	Alkaline EDTA pH \geq 12	Chopped bipolar sawtooth	>99.9
Ga-67	Zn-68	Diluted HCl pH \geq 2.5	Chopped cathodic sine	>99.9
In-111	Cd-112	Ammonia PH \gg 7	Asymmetric square wave	>99.9
Pd-103	Rh-103	Sulphuric acid 1 < pH < 2	DC voltage	40–70

Apart from rhodium, AC waveforms resulting in a quantitative current yield are used. For rhodium, electroplating to depletion (>98% of rhodium metal deposited) is recommended.

4.3. QUALITY CONTROL OF ELECTROPLATED TARGETS

As the greater part of industrial demands can be met by proper design of the plating set-up and associated electronics, quality control mainly focuses on the physicochemical requirements reported in Section 4.2.1. The following tests should be used as quality control criteria:

- (a) The CMP of the plating and recovery solutions may affect the radionuclidic and/or chemical purity of the radionuclide bulk obtained after irradiation and processing of the target. The chemical purity should be determined using selective multi-element analytical techniques such as stripping voltammetry (SV) or induced coupled plasma spectrometry (ICP). A CMP of 99.9% should be obtained. It should be noted that the CE of the plating process might be considered to be a co-evaluation of the CMP. Co-deposition and/or hydrogen evolution (in general having a negative effect on the quality of the deposit) may be expected whenever $CE < 100\%$. Therefore the development of CCE electroplating technology showing a $CE < 99\%$ is strongly recommended, in which case simple weighing of the target allows on-line CE control.
- (b) The SAG is important as it influences both the local effective thickness of the target layer and the local particle scatter probability. Poor and non-reproducible SAG will result in an enhanced scatter of the radionuclide

production yield. To evaluate the granulometry, an ‘acceptable’ reference should be defined. The surface grain size distribution determined by microscopy or SEM at a given magnification should be compared with that of a target of interest. As a criterion, ‘comparable with’ can be used.

- (c) The homogeneity of the target layer (HTL) can be determined by micrometry or by transmission or reflection measurements. As the SAG, the HTL may strongly affect the reproducibility of the production yield. An acceptable value of HTL should be an SD of 5% on the set of twelve measurements spread all over the target surface area.
- (d) The TST reveals the incorporation of organic plating additives or the presence of occlusions and/or vacuoles. A practical TST involves heating up of the target in the air or in a vacuum at a temperature well below (at least 30–40°C) the melting point of the target metal (or of the target carrier material if it shows a lower melting point), followed by quick submersion of the hot target in cold (15°C) water and subsequent multiple bending of the carrier. No cracks or craters should be observed and no peeling off of the target layer should occur.

It should be noted that apart from the thermal shock, all quality control tests can be performed on-line. The TST should be considered to be a quality control test on the plating technology and should be tried with natural target material.

4.4. OPTIMIZATION OF THE COOLING AND IRRADIATION MODE

As stated previously, the efficiency of the cooling and the particle beam quality are as important as the quality of the electroplated target layer when high beam current ($I > 200 \mu\text{A}$) irradiations are intended. The efficiency of the cooling is determined by both the target carrier geometry and the flow rate of the coolant while the beam quality deals with the current density distribution on the irradiated surface area.

4.4.1. Optimization of the cooling

In view of heat generation and cooling, an electroplated target can be considered to be a three layer system. This includes a homogeneous metal (M) target layer of thickness a and surface area S in which the radionuclide of interest is produced, a copper target carrier layer of thickness b in which the impinging particles dissipate their remaining energy and a carrier layer of thickness c required to ensure sufficient mechanical strength to withstand the

coolant pressure. Assuming a homogeneous beam current density and applying Poisson's equation to layers a and b , Laplace's equation to layer c and Newton's equation to the carrier-coolant interface, the temperature profile over the multilayer system and the maximum temperature of the target layer T_m can be calculated:

$$T_m - T_w = \left(\frac{q_M}{2 \cdot K_M} \cdot \frac{a}{S} \right) + \left(\frac{q}{K_{Cu}} \cdot \frac{b}{S} \right) + \left(\frac{q_{Cu}}{2 \cdot K_{Cu}} \cdot \frac{b}{S} \right) + \left(\frac{q_T}{K_{Cu}} \cdot \frac{c}{S} \right) + \frac{q_T}{h \cdot S} \quad (1)$$

In this equation T_w is the coolant temperature, K_M and K_{Cu} the thermal conductivity coefficients of the target material and of the copper carrier, h the convective heat transfer coefficient at the copper carrier-coolant interface and q_m and q_{Cu} the heat generated in the target and copper layers respectively. For a given particle entrance energy, the latter can be evaluated from the mean energy loss per unit path length, the physical thickness of the layers and the beam target angle. For turbulent flow, the value of h can be calculated from the Dittius-Boelter equation: its magnitude increases as the flow rate increases. Equation (1) can be rewritten as a function of the applied beam current I :

$$T_m - T_w = \left(\frac{k_1 \cdot I}{K_M} \cdot \frac{a}{S} \right) + \left(\frac{k_2 \cdot I}{K_{Cu}} \cdot \frac{b}{S} \right) + \left(\frac{k_3 \cdot I}{K_{Cu}} \cdot \frac{b}{S} \right) + \left(\frac{k_4 \cdot I}{K_{Cu}} \cdot \frac{c}{S} \right) + \left(\frac{k_4 \cdot I}{h \cdot S} \right) \quad (2)$$

where k_1 , k_2 , k_3 and k_4 are constants. By substituting the maximum allowable TST temperature discussed in Section 4.3 for T_m in Eq. (2), the maximum beam current burden $I_{T,m}$ of the target can be obtained. The maximum beam current density on target is then:

$$i_{m,T} = I_{T,m} \cdot S \quad (3)$$

As an example, proton irradiation of a ^{203}Tl target may be used. For 28.5 MeV bombardment of an 80 μm thick thallium layer deposited on a 2 mm thick flat copper backing, a coolant channel showing a rectangular cross-sectional area of 0.3 cm^2 (length 12 mm, width 2.5 mm) through which a coolant flow rate of 50 L/min is forced and with $T_m = 270^\circ\text{C}$ and $T_w = 21^\circ\text{C}$, the maximum allowable current density on target equals:

$$i_{m,T} = 0.5 \mu\text{A}/\text{mm}^2 \quad (4)$$

Equation (4) applies for a 90° beam/target angle geometry. When this angle is reduced to ϑ ($\ll 90^\circ$), the maximum current density at the exit plane of a cylindrical collimator amounts to:

$$i_{m,C} = i_{m,T}/\sin(\vartheta) \quad (5)$$

For the given example and with $\vartheta = 6^\circ$, $i_{m,C} = 5 \mu\text{A}/\text{mm}^2$. When the thickness of the flat copper carrier is reduced to 0.5 mm, $i_{m,C} = 6.3 \mu\text{A}/\text{mm}^2$. Doubling the coolant flow rate to 100 L/min then results in an $i_{m,C} = 9.7 \mu\text{A}/\text{mm}^2$, i.e. the maximum target current burden will be almost doubled.

From Eq. (2) it follows that for a given target the maximum allowable target current can be increased by:

- (a) Reducing the thickness (a) of the target layer to a minimum. To take full advantage of the excitation function this implies an appropriate reduction of the beam/target angle. To avoid unwanted scatter of the impinging particles this angle should not be less than 6° .
- (b) Increasing the irradiated surface area (S), requiring redesign of the collimator geometry and retuning of the beam optics parameters and/or reducing the beam/target angle.
- (c) Reducing the thickness ($b + c$) of the target carrier to a minimum value. This can be done by omitting cooling fins and reducing the thickness of the copper layer so that it can withstand the coolant fluid pressure. Experiments show that a one mm copper plate with a surface area of about 10 cm^2 can be pressurized to 45 bar^1 before it cracks. Calculations show that reduction of the thickness of the copper support from 2 to 0.5 mm allows an increase of the maximum current of about 30%. For safety reasons an 0.5 mm thick flat copper target carrier pressurized to 30 bar should be recommended.
- (d) Increasing the coolant flow rate, thus increasing the convective heat transfer coefficient h . For a coolant channel showing a normal rectangular cross-sectional area of 0.3 cm^2 and a width of 2.5 mm the value of h nearly doubles when the mean linear flow rate is changed from 2600 cm/s ($\approx 50 \text{ L}/\text{min}$) to 5200 cm/s ($\approx 100 \text{ L}/\text{min}$). To avoid excessive coolant pressure being required to obtain this flow rate, the hydrodynamic resistance at the inlet and outlet of the coolant channel should be reduced to a minimum. This can be done by substituting the frequently used rectangular inlet and outlet/coolant channel geometry for a small angle one.

¹ 1 bar = $1.00 \times 10^5 \text{ Pa}$.

4.4.2. Optimization of the irradiation mode: circular wobbling of the beam

In practice a Gaussian current density distribution at the exit of a cylindrical collimator is assumed:

$$i = i_{m,C} \cdot e^{-a^2 r^2} \quad (6)$$

with $i_{m,C}$ the maximum axial current density at the collimator exit plane, i the current density at a distance r from the axis and a^2 the Gaussian exponent. For a given cylindrical collimator with radius R , the latter can be easily evaluated from the collimator (I_C) and target (I_T) currents:

$$a^2 = \frac{1}{R^2} * \ln\left(\frac{I_T + I_C}{I_C}\right) \quad (7)$$

The maximum allowable target current $I_{T,m}$ can be calculated from Eq. (8):

$$I_{T,m} = \frac{\pi * i_{m,C}}{a^2} \left(1 - e^{-a^2 R^2}\right) \quad (8)$$

The corresponding collimator current is equal to:

$$I_{C,m} = \frac{\pi * i_{m,C}}{a^2} \cdot e^{-a^2 R^2} \quad (9)$$

and the associated total beam current $I_{B,m}$ to:

$$I_{B,m} = \frac{\pi * i_{m,C}}{a^2} \quad (10)$$

On condition that the maximum allowable collimator current burden is not the limiting factor, Eq. (9) allows calculation of the Gaussian exponent required to fully exploit the maximum beam current output (I_{acc}) of a given accelerator:

$$a^2 = \frac{\pi * i_{m,C}}{I_{acc}} \quad (11)$$

The beam transport parameters should then be adjusted so that introduction in Eq. (7) of the measured values I_C and I_T results in the same

value of a^2 as that calculated from Eq. (10). For an accelerator with $I_{\text{acc}} = 400 \mu\text{A}$, $R = 5 \text{ mm}$ and a targetry/coolant system allowing $i_{\text{m,C}} = 5 \mu\text{A}/\text{mm}^2$, a value of $a^2 = 0.04 \text{ mm}^2$ is calculated. This corresponds with $I_{\text{T,m}} = 247 \mu\text{A}$ and $I_{\text{C,m}} = 153 \mu\text{A}$, i.e. about 38% of the beam current is absorbed by the collimator.

In general, for most of the commercially available collimators the maximum current burden is limited to $100 \mu\text{A}$. To benefit fully from I_{acc} , circular wobbling should be applied. Therefore the beam is symmetrically centred (using a four quadrant collimator) on the axis of the collimator and the beam optics parameters adjusted so that an appropriate value of a^2 is obtained. Next, the beam is shifted out of the axis over a distance R_{W} (the wobblers' radius) and forced to rotate with a given period (the wobblers' period T_{W}) about the axis. The result is a more or less homogeneous current density distribution with a lower maximum current density as compared with the non-wobbled Gaussian one, a higher maximum allowable beam current on target and a limited current load on the collimator.

The calculations of the maximum mean current distribution density and of the total beam current on target $I_{\text{T,m}}$ cannot be done analytically, numeric integration is required. This can be done for the ^{203}Tl target mentioned previously using the CIRCWOB code, a Qbasic algorithm developed at the Cyclotron Department of the Vrije Universiteit Brussel, Belgium. As input parameters i_{M} , the maximum current density of the non-wobbled Gaussian beam, a^2 the Gaussian exponent, R_{W} the wobbler radius and its increment ΔR_{W} and T_{W} the wobbler period are used. For a maximum current density of $i_{\text{m,C}} = 5 \mu\text{A}/\text{mm}^2$ at the exit plane of a cylindrical collimator with radius $R = 5 \text{ mm}$, with $\Delta R_{\text{W}} = 0.01 \text{ mm}$, $T_{\text{W}} = 0.02 \text{ s}$ and a maximum accelerator beam current $I_{\text{acc}} = 392 \mu\text{A}$, a typical output is represented in Table 2.

TABLE 2. OUTPUT OF THE CIRCWOB.EXE CODE FOR
 $i_{\text{m,C}} = 5 \mu\text{A}/\text{mm}^2$

i_{M} ($\mu\text{A}/\text{mm}^2$)	a^2 (mm^2)	R_{W} (mm)	$i_{\text{m,C}}$ ($\mu\text{A}/\text{mm}^2$)	$I_{\text{T,m}}$ (μA)	$I_{\text{C,m}}$ (μA)	I_{acc} (μA)	μ_{B} (%)
10	0.080	2.94	5	277	115	392	70.66
15	0.120	3.04	5	295	97	392	75.25
20	0.160	3.16	5	310	82	392	79.1
25	0.200	3.37	5	313	79	392	79.8
30	0.240	3.61	5	307	85	392	78.3
35	0.280	3.85	5	297	95	392	75.8
40	0.320	4.08	5	283	109	392	72.2

From these data it is obvious that an accelerator beam output current showing a Gaussian distribution with $a^2 = 0.200 \text{ mm}^2$ and a maximum beam current density of $i_M = 25 \text{ }\mu\text{A}/\text{mm}^2$ without wobbling should be presented to the wobbling device operating at a W_R of 3.37 mm. As only $5 \text{ }\mu\text{A}/\text{mm}^2$ current density at the collimator output plane (and $0.5 \text{ }\mu\text{A}/\text{mm}^2$ on target) is tolerated, the beam optics parameters should be adjusted with a bare copper target carrier prior to the thallium target irradiation.

From Eq. (6) it follows that for $a^2 = 0.200 \text{ mm}^2$, $R^2 = 25 \text{ mm}^2$ and $I_T + I_C = I_{\text{acc}} = 392 \text{ }\mu\text{A}$, this beam tuning should result in a collimator current $I_C = 2.6 \text{ }\mu\text{A}$ and $I_T = 389.4 \text{ }\mu\text{A}$ with $0.64 \text{ }\mu\text{A}$ on each quadrant of the collimator. Upon inserting the target Faraday cup in the beam path, the thallium target should be mounted and the wobblers radius adjusted so that the collimator current equals $79 \text{ }\mu\text{A}$, whereupon the Faraday cup should be removed from the beam path. It should be noted that the maximum collimator current burden is much lower than $100 \text{ }\mu\text{A}$.

From the foregoing it follows that if no wobbling of the beam is applied, the optimization of the target irradiation mode may involve:

- (a) Adjustment (decrease) of the Gaussian exponent (a^2) implying that the beam optics parameters should be tuned so that a broad spatial current density distribution at the collimator exit plane is obtained, the maximum of which should never exceed $i_{m,C}$. This means that the collimator design should allow a high current burden to fully exploit the beam intensity performances of present day ($I_{\text{acc}} = 400 \text{ }\mu\text{A}$) accelerators.
- (b) Proper design of the collimator cooling and geometry so that the centreing of the spatial beam current distribution can be monitored and adjusted accordingly. A simple four quadrant well-cooled collimator may satisfy this demand.

When circular wobbling is applied, the collimator current burden may be greatly reduced and the maximum allowable target current maximized. Apart from the items cited above, this requires:

- (1) Installation on an inexpensive three-coil wobbling system operating at the mains frequency in the target beam line;
- (2) Formulation of an appropriate startup procedure involving bare target carrier irradiation before target bombardment that should be executed rigorously by every cyclotron operator.

4.5. CONCLUSIONS OF SECTION 4

The development of a plating technology for the production of high quality solid targets should be accompanied by proper design of the target carrier and collimator geometry and the cooling of both, and by optimization of the irradiation mode.

To meet the major physicochemical quality requirements (chemical purity, homogeneity, SAG, thermal stability), target plating should preferably be done by time controlled CCE at room temperature with 100% plating CE. This is feasible only when the composition of the plating solution (metal concentration, pH, complexing agent, surfactant, depolarizers and stress reducing agent) and the applied plating current density are adjusted so that gas evolution at the cathode, concentration polarization, dendrite formation and incorporation of bath additives are avoided. In this context plating from alkaline solutions containing a complexing agent showing a low stability constant ($K_{\text{stab}} = 6-7$) with the target metal should be recommended and, if oxygen evolution is to be avoided, hydrazine hydrate should be used as the anodic depolarizer. As a non-incorporated surfactant BRIJ-35 can be tried. In addition the plating vessel geometry should result in a homogeneous electrical field all over the plating area and efficient stirring should prevent local concentration gradients. A cylindrical plating vessel fitted with a single axial grounded anode wire, with a multiple of four windows holding the target carriers in the vertical wall and with a perforated cylindrical bi-directional electromechanical stirrer surrounding the anode satisfies these demands. Finally, ultimate dendrite suppression can be obtained by applying an AC waveform of appropriate shape and frequency.

Four criteria allow the quality of a solid target to be estimated. The CMP (CMP \geq 99.9%) determined by multielement analytical techniques (SV, ICP) hinders unexpected chemical and radionuclidic impurity of the final product. The SAG (SAG, comparable with reference) as estimated by optical techniques (SEM), limits local particle scattering and subsequent dispersion on the production yield. As SEM, the HTL (HTL, standard deviation on the thickness $SD \leq 5\%$ for $n = 12$) limits the scatter on the yield. Finally, the TST (one hour heating at $T^\circ = \text{melting point} - 40^\circ\text{C}$, submersion in cold water, multiple bending, no cracks, no craters, no peeling off) warrants no incorporation of plating additives and avoids losses of enriched material during the irradiation and subsequent unreproducible production yields.

Target cooling can be optimized by calculation of the maximum allowable target current burden (and target current density), taking into account the maximum allowable target layer temperature as determined by the TST. Postulating a three layer target model this can be done by application of

classical heat transfer equations. Optimum coolant channel geometry should show a narrow (width = 2–3 mm) normal rectangular cross-section with a thickness of the flat target layer (copper) support plate (no fins) of about 1–2 mm and a coolant flow rate ensuring full turbulent flow ($v > 2500$ cm/s). To avoid the necessity of excessive coolant pressure, the hydrodynamic resistance (and subsequent pressure drops) at the inlet and outlet of the coolant channel should be reduced by substituting the frequently used rectangular geometry for a small angled one.

Optimization of the irradiation mode depends on whether or not beam wobbling is applied. Assuming a Gaussian current density distribution, which should be checked by an appropriate beam diagnostic device, a simple four quadrant collimator design may satisfy. Bearing in mind the maximum allowable current density on target as calculated from the three layer model, a maximum allowable target beam current can be calculated easily. Beam optics parameters may then be set using the target and collimator current readings only. In general this entails a high collimator beam current and requires well adapted cooling. Moreover it results in high activation of the collimator, entailing a high radiation dose rate during maintenance. Therefore circular beam wobbling is preferable. Apart from a diagnostic beam device, it requires the installation of an inexpensive three coil wobbling system and the formulation of an appropriate startup procedure that should be executed rigorously by the cyclotron operator.

5. RESULTS OF THE WORK ON TARGET FABRICATION FOR ^{123}I AND ^{124}I PRODUCTION

5.1. INTRODUCTION

Iodine radioisotopes have played a fundamental role in biomedical research and clinical nuclear medicine. The wide variety of decay characteristics and half-lives available with these isotopes have made them useful for both diagnostic imaging and therapy. Due to its favourable and almost ideal physical properties, such as high abundance of low energy photons (83%, 159 keV) emitted after 100% electron capture decay with a half-life of 13.2 h, ^{123}I has been a staple in diagnostic imaging by single photon emission computerized tomography (SPECT) (thyroid uptake, cardiological, neurological and oncological diseases) and continues to be a valuable isotope

for research in receptor binding assay and metabolism [2, 3]. Iodine-124 (23% positron emission, half-life 4.2 d) has attractive properties for use in diagnostic PET imaging with monoclonal antibodies, and has potential for therapy [4].

For the production of ^{123}I , gaseous, liquid and solid targets can be used depending on the energy of the cyclotron and the availability of the enriched target material, especially ^{124}Xe . Each type of target has its own advantages and disadvantages [5]. High purity ^{123}I production, involving the $^{124}\text{Xe}(p,pn)^{123}\text{Xe} \rightarrow ^{123}\text{I}$ and $^{124}\text{Xe}(p,2n)^{123}\text{Cs} \rightarrow ^{123}\text{Xe} \rightarrow ^{123}\text{I}$ threshold reactions, is becoming more and more popular as it allows an easy recovery of the iodine. The main drawbacks are the very expensive and sophisticated targetry set-up and the high price of ^{124}Xe [3]. Molten NaI targets require bombardment with higher energy particles of more than 50 MeV to make use of the $^{127}\text{I}(p,5n)^{123}\text{Xe} \rightarrow ^{123}\text{I}$ reaction [3].

One of the most common target materials for the production of ^{123}I is enriched $^{122,123,124}\text{Te}$, either as a metal or as tellurium dioxide [2, 5, 6]. Powdered tellurium, often mixed with aluminium powder to increase the heat transfer characteristics, has been widely used for targets. The common problem with these targets is the melting of the elemental tellurium (MP 452°C) and consequent loss of radioiodine from the matrix. The use of $^{122,123,124}\text{TeO}_2$ showing a higher melting point (733°C) may reduce this loss. Tellurium oxide is usually melted into the cavity of a small target plate. Small amounts of aluminium oxide may be added to aid in adhesion of the oxide to the surface of the plate. Higher beam currents can be tolerated when the tellurium target material is electroplated on a suitably large area target carrier and when a small beam/target angle irradiation is performed in optimum cooling conditions.

Up to now the targetry involved in the production of ^{124}I has been limited to solid targets. The targets are either elemental tellurium or tellurium dioxide and are identical to those used for the production of ^{123}I from tellurium [7].

From the above it is obvious that large area electroplated tellurium targets are attractive tools for higher beam current irradiation as long as the deposits are smooth, homogeneous and free of plating bath constituents. Tellurium coatings being of no practical importance to industry, appropriate plating solutions are not commercially available. To avoid the poor quality target layers frequently obtained when plating procedures reported in the literature are applied, a new plating technology involving CCE multi-target electroplating from alkaline medium and showing 100% plating CE was developed. By way of comparison, TeO_2 solid targetry is also discussed.

5.2. TARGET FABRICATION

5.2.1. Electroplated Te targets

Tellurium targets are prepared by DC CCE of the metal from alkaline plating solutions. Copper plates coated with nickel are used as target carriers.

Pretreatment of the copper carrier

To ensure good adhesion of the target material to the target carrier, the latter is coated with a 50 μm nickel layer before tellurium plating. Following is the nickel plating procedure:

- (1) Dissolve 500 g of $\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$ in 1000 mL of de-ionized water;
- (2) Clean the copper carriers by rubbing with sandpaper (320–500 mesh grade), followed by rinsing with de-ionized water;
- (3) Immerse the copper plate (cathode) and a platinum wire (anode) in a 1000 mL beaker containing the nickel plating solution;
- (4) Connect the electrode set to a DC power supply, switch on the power and adjust the current to 400–800 mA;
- (5) Plate out the nickel for 3 min, then remove the electrode set from the solution;
- (6) Rinse with water and acetone and air dry the target carrier.

Preparation of the tellurium plating solution

The plating solution is prepared from enriched $^{124}\text{TeO}_2$ (abundance >99%) that is dissolved in alkaline medium:

- (1) Introduce 3.0 g $^{124}\text{TeO}_2$, 7.5 g KOH and about 250 mL de-ionized water in a 400 mL beaker;
- (2) Introduce a magnetic stirring bar and homogenize for 10 min;
- (3) Remove any residual particle by filtration through a 0.45 μm filter;
- (4) Adjust the volume to 250 mL.

Alternatively, the plating solution can be prepared from recovered ^{124}Te metal. In this case 2.4 g of tellurium are dissolved in a few milliliters of 8N HNO_3 . The solution is heated to boiling until evolution of nitrogen oxides ceases, whereupon KOH and water are added.

Simultaneous plating of four 20 μm target layers (surface area 6.6 cm^2)

The plating vessel is a hollow Perspex cylinder fitted with an axial one mm platinum anode wire mounted in the bottom. Four symmetrical windows (surface area 6.6 cm^2) in the vertical wall of the cylinder allow the introduction and positioning of four pretreated target carriers with a thickness of 1.5 cm. Each slot is provided with a window fitted with an O ring, the geometrical shape of which determines the electrodepositing area. Liquid tightness is realized by stainless steel mechanical pestles mounted in a PVC ring surrounding the plating vessel and by pressing the copper carriers against the O ring. The external PVC ring is also fitted with four supporting pins to hold a motor stirrer combination in position. The stirrer is a hollow perforated Poly Oxymethylene (polyacetol) cylinder surrounding the platinum anode and is mounted on the axis of the DC motor. The stirrer rotation speed is set at 1200 rev./min. The plating is done at room temperature.

Constant current electrolysis is used as the plating method. Therefore, the output of a single adjustable DC generator is connected to four V/I converters/ current boosters, the outputs of which are connected to the Ni coated cathodes.

Use the following procedure:

- (1) Introduce the four Ni coated copper carriers into the four windows of the plating vessel and connect them to the four booster outputs. Transfer the freshly prepared plating solution into the plating vessel.
- (2) Introduce the electromechanical stirrer, switch on the motor and adjust the rotation speed to 1200 rev./min.
- (3) Start the electrolysis by connecting the platinum anode to the ground; adjust the generator voltage so that a 75 mA current per target is obtained.
- (4) Continue plating for 1.1 h.
- (5) Stop the stirrer, remove it from the solution and rinse. The rinse water should be added to a recovery bulk. Transfer the plating solution to the beaker and switch off the voltage.
- (6) Remove the targets and rinse them abundantly with de-ionized water. Air dry the targets and store them in a vacuum desiccator.

A mean weight of 90 ± 9 mg of enriched tellurium is deposited per target. This means that the average plating CE is about 100%. The plating solution can be reused as many as four times as the DR should not be higher than 60% to ensure a good quality deposit.

Recovery of enriched ^{124}Te

The enriched material from a set of partially depleted plating solutions (i.e. about 0.95 g Te per plating solution) or from combined recovery/rinsing solutions obtained after solubilization of irradiated targets and separation of the iodine (90 mg Te per run) is recovered as solid tellurium metal. Therefore, the enriched material is solved as tellurate in a mixture of hydrochloric acid and hydrogen peroxide, followed by the partial reduction to tellurite by hydrogen bromide. Quantitative precipitation as metallic tellurium is achieved by reduction with hydrazine and sodium sulphite.

Use the following procedure:

- (1) Combine as many as four partially depleted plating solutions and evaporate to near dryness, then add 200 mL of a $\text{HCl}/\text{H}_2\text{O}_2(30\%)$ 3:1 mixture. Alternatively about 4–5 g of tellurium obtained from target processing may be introduced into the same mixture.
- (2) Evaporate the solution until the volume is reduced to about 100 mL — a clear yellow solution should be obtained.
- (3) Filter the warm solution through an 0.45 μm glass filter to remove any residual particle.
- (4) Add 40 mL of HBr (48%) to reduce the tellurate to tellurite and heat until the volume again equals about 100 mL.
- (5) Stepwise, while vigorously stirring, add solid $\text{NH}_2\text{NH}_2 \cdot 2\text{HCl}$ until tellurium starts to precipitate. Then introduce 5 g Na_2SO_3 , heat and stir for 15 more min.
- (6) Filter through an 0.45 μm glass filter and wash the precipitate abundantly with water and acetone.
- (7) Dry the precipitate for 3 h at 300°C and weigh.

The recovered material is stored as such until it is reused to prepare a new plating solution.

5.2.2. Oxide targets

The target is prepared by melting isotopically enriched (^{123}Te or ^{124}Te > 90% abundance) TeO_2 onto a small platinum plate. The chemical purity of the starting material is important not only for the chemical and radionuclidic purity of the radionuclides produced, but also for the preparation of the target itself since the adhesion of tellurium dioxide to the platinum backing plate greatly depends on its purity. Furthermore, the platinum surface should not be smooth, but should be scratched heavily with a scalpel or lancet before

preparation of the target. The target quality also depends on the amount of tellurium dioxide powder used. If this amount is small, a small drop will be formed upon heating and the TeO_2 will be attached only slightly to the bottom of the plate.

When a rectangular profile of the plate cavity is used and a sufficient amount of powder is introduced, a pronounced non-uniformity of the target material is observed with an empty centre and thick layer along the edges. As it is very desirable to have a uniform target thickness, up to 10% Al_2O_3 is often used to ensure acceptable adhesion to the carrier and uniformity of the glassy layer. Finally, the cooling rate strongly affects the ultimate quality of the layer. If upon melting the target is cooled too fast the TeO_2 layer may be destroyed. Therefore slow cooling is required.

Preparation of a tellurium dioxide target (surface area 0.8 cm²)

- (1) Prepare a circular platinum plate by pressing a 2.2 cm × 2.2 cm platinum foil (thickness 0.3 mm) in an appropriate deep draw mould/stamp combination. The inner diameter of the cavity is one cm, its depth equals 0.8 mm.
- (2) Heavily scratch the bottom of the cavity with a scalpel or lancet so that a rough surface is obtained.
- (3) Weigh 250–280 mg of TeO_2 in the plate and mount it perfectly horizontally in a cold oven.
- (4) Switch on the furnace power supply and heat to 750°C. This takes about 15 min.
- (5) Heat for five more minutes at this temperature, then switch off the power supply and allow the oven to cool slowly.

Alternatively, the starting material may be metallic enriched tellurium. This is converted to TeO_2 by dissolving the metal in hot 60% HNO_3 . Upon evaporation to dryness, the residue is annealed in an oven at 450°C over four h. The stoichiometry of the resulting tellurium oxide corresponds to TeO_2 and the losses of enriched material during the process are negligible.

Recovery of enriched tellurium dioxide

Enriched material from irradiated TeO_2 targets is recovered by means of a multi-stage process. It involves vacuum distillation of Te and TeO_2 , acid dissolution and ultimate precipitation as metallic tellurium.

In the first stage, a small amount (as much as 200 mg) of contaminated TeO_2 is vacuum distilled ($p = 3.10^{-3}$ mm Hg, $T = 770\text{--}800^\circ\text{C}$) in a quartz

apparatus for one hour. Next, hot concentrated hydrochloric acid is added to the non-volatile residue and, with 0.45 μm filtration, a clear acid solution acid is obtained. From the latter the tellurium is precipitated as metal, using the procedure described in Section 5.2.1. Finally, the tellurium metal is vacuum-distilled and the collected material transformed into tellurium dioxide as described in Section 5.2.1. The overall losses during the recovery procedure equal 3–4%.

5.3. TARGET IRRADIATION

The feasibility of using high beam currents for radioisotope production depends on several factors. The most important of these are the nature (melting point, thermal conductivity coefficient) of the target material, the efficiency of the target cooling and the quality of the particle beam, i.e. the current density distribution on the target. Each of these parameters will influence both electroplated and oxide targets for the production of radioiodine.

5.3.1. Plated target irradiation

In typical experiments, ^{123}I is produced by bombarding enriched ^{124}Te targets (90 mg \pm 10%) in a 1° beam/target geometry with 26 MeV internal proton beams using the $^{124}\text{Te}(p,2n)^{123}\text{I}$ nuclear threshold reaction.

The applied current range is 18–45 μA , the irradiation time amounts to 2.0–2.75 h. A substantial increase in iodine yield is measured when the current rises from 18 to 24 μA .

Though no losses of enriched tellurium target material are found in the current interval of 24–45 μA , currents higher than 24 μA should not be applied as a decreasing yield of iodine is observed. This decrease in yield must be attributed to the high temperature generated from the high beam current, which causes evaporation of the easily volatilized ^{123}I in a vacuum. Therefore further optimization of the beam position, the beam current and power density distribution on the target and of the efficiency of the cooling system is required. For internal targets, this implies optimization of the harmonic coil settings. For external beam irradiation, reduction of the beam/target angle, increase of the irradiated surface area, linear scanning or circular wobbling may be tried.

5.3.2. Oxide target

In most cases iodine radionuclides (^{123}I , ^{124}I or ^{125}I) are produced by proton irradiation of tellurium dioxide deposited on a platinum backing plate. The heat conductivity of tellurium dioxide being rather poor, it can be expected that the targets will not withstand high beam currents. As energy losses in the target material depend on the proton energy, higher beam currents can be applied with higher energy particles. Typically, ^{123}I and ^{124}I are produced using proton entrance energy of about 15 MeV. Water cooling of the backing plate and helium cooling of the tellurium oxide layer are applied. With a tellurium dioxide thickness of about 250 mg/cm^2 the maximum allowable wobbled beam current is limited to $6\text{--}8 \mu\text{A/cm}^2$ that can be applied at least during 12 to 14 h in a 90° beam/target angle geometry.

5.4. TARGET PROCESSING

5.4.1. Electroplated target

To avoid losses of the carrier free iodine radionuclide, the irradiated target is dissolved in oxidizing alkaline medium, followed by reduction of the enriched tellurium to metal and of the iodine to the I^{-1} state by aluminium powder. The iodine is purified by cation exchange chromatography.

The irradiated target layer is dissolved at room temperature in 10 mL of a mixture of NaOH, H_2O_2 and water (1.0 mL 5.0N NaOH + 3 mL H_2O_2 30% + 6 mL Milli-Q water). After 20–30 min, the solution is transferred into a 250 mL quartz reaction flask containing 160 mg of aluminium powder. Upon washing the target carrier with 2 times 5 mL of the mixture, the tube is heated to boiling until the reaction is completed. Next air is bubbled into the solution for five min, followed by bubbling CO_2 for an additional five min to allow quantitative precipitation of tellurium and $\text{Al}(\text{OH})_3$. The solution is then carefully filtered through an $0.45 \mu\text{m}$ glass filter and an in-line AG50W-X8(H^+ , 100–200 mesh grade, 1 cm \times 5 cm) column. When more than 5% of the iodine activity remains on the column, the latter is washed with 5 mL Milli-Q water. The eluate is collected into a pre-weighted serum vial. If necessary, the pH of the ^{123}I bulk is adjusted in the range between 5 and 7 with 0.01N NaOH. The overall yield of the chemical processing is higher than 95%.

5.4.2. Oxide targets

The dry distillation method is used to extract carrier free iodine. Therefore the irradiated sample is introduced into a quartz tube horizontally mounted in a cylindrical mini-oven and the iodine trap vial is filled with 0.01N NaOH. The air carrier flow is started and the power supply of both the furnace and the heating element wrapped around the narrow quartz outlet tube are switched on.

Liberation of iodine from the target starts at a temperature of about 620°C, at a distillation rate governed by the diffusion of iodine from the tellurium oxide bulk to its surface. Between 710 and 740°C (MP TeO, 733°C), an iodine vapour outburst takes place. Experimentally it has been found that periodic melting and solidification from this point on results in a 98% recovery of the radioiodine. Therefore, 10 min after the start of the distillation, the furnace power supply is switched on and off so that the temperature oscillates between 700 and 740°C. Losses of TeO₂ are limited to less than 0.2%.

5.5. TARGET QUALITY CONTROL

5.5.1. Electroplated Te targets

The new electroplating technology developed allows the production of high quality enriched ¹²⁴Te targets. Microscopic inspection shows a smooth, dendrite free surface area with excellent granulometry. The targets withstand a 150°C TST.

5.5.2. Tellurium dioxide targets

Upon preparation of the targets, the presence of impurities in the raw target material will result in a non-homogeneous, coloured layer. Therefore, visual inspection is done after preparation. Acceptable targets should be glassy and show a flat surface. Next, the inspected target is irradiated for one hour only, followed by a second visual control and determination of the yield and of the radionuclidic purity of the iodine radionuclide of interest. If within acceptable limits, the target is submitted to long time irradiation for production.

5.5.3. Iodine batch quality control

Iodine batches produced through irradiation of solid Te and TeO₂ targets are submitted to chemical purity tests on aluminium and tellurium. Use is made of emission spectrometry or colorimetry. The iodine batches produced through both targets meet all criteria of the pharmacopoeia. Typical radionuclidic specifications of a ¹²³I batch obtained with plated ¹²⁴Te (99%) targets are as follows: ¹²³I, 96.4%; ¹²⁴I, 3.4%; ^{126,130}I, 0.2%. The chemical impurities determined by colorimetric spot tests are typically: Te < 0.5 µg/mCi;² Al < 0.25 µg/mCi. For TeO₂ targets the radionuclidic purity amounts to 99.3% for ¹²³I and 99.97% for ¹²⁴I.

5.6. CONCLUSIONS OF SECTION 5

The production yield of ¹²³I and ¹²⁴I radionuclides by solid target irradiation strongly depends on the quality of the target. To fabricate good quality electroplated metallic tellurium layers deposited on Ni coated copper carriers, a new DC CCE plating technology was developed. The plating is done using a home-made cylindrical plating vessel fitted with 4 (6.6 cm²) windows holding the carriers (cathodes) and a single axial grounded platinum anode wire. Under vigorous stirring by means of a perforated cylindrical electromechanical stirrer (1200 rev./min), 4 smooth, homogeneous and dense 20 µm target layers are prepared simultaneously within one hour when an alkaline (0.5N KOH) plating bath is used and the plating current density is adjusted to 11.4 mA/cm². As the plating CE is nearly 100%, time controlled plating is feasible. The layers withstand a 150°C TST and tolerate an internal beam current irradiation (1° beam/target angle) of as much as 45 µA with 26 MeV protons — the ¹²⁴Te(p,2n)¹²³I is used without losses of enriched material. For reasons most probably associated with the non-homogeneous beam current density distribution on target and poor cooling efficiency, the practical beam current burden in the Cyclone-30 should be limited to 24 µA. In this case, the yield for ¹²³I equals 16.61 mCi.µAh⁻¹ and per batch about 250 mCi is produced when the irradiation time is limited to 2.5 h. This implies an increase of about 22% per batch as compared with that obtained with targets plated according to procedures available from the literature. Further optimization of the beam current distribution and/or the cooling efficiency for

² 1 Ci = 3.70 × 10¹⁰ Bq.

internal target irradiation, or the use of the plated targets in wobbled external beam irradiation of well cooled targets, may improve the yield per batch.

For the tellurium dioxide targets, a yield of about 270 mCi of ^{123}I per batch is obtained when the ^{123}Te target is irradiated for 15 h in a 90° beam/target angle geometry with $6\ \mu\text{A}$ 15 MeV protons. In this case the $^{123}\text{Te}(p,n)^{123}\text{I}$ is utilized. Linear extrapolation to an irradiation with $24\ \mu\text{A}$ should give 1080 mCi per batch.

As compared with the solid ^{124}Te target yield of 1500 mCi ($24\ \mu\text{A}$, 15 h) this is in agreement with the magnitude of the maximum cross-section values as read from the excitation curves for the (p,n) and (p,2n) reactions, respectively. For the tellurium dioxide target, experiments show that enlargement of the target surface area is the most promising way to increase the yield of iodine per batch. An on-line beam scanning device for beam profile measurements and wobbling should be very advantageous for the reliable production of ^{123}I or ^{124}I using this target. The purity of that target material is also critical in obtaining a good reliable yield. Quality control procedures must be carried out on the target before final irradiation.

6. RESULTS OF THE WORK ON TARGET FABRICATION FOR ^{103}Pd PRODUCTION

6.1. INTRODUCTION

The suitability of a given radionuclide for brachytherapy is determined by its half-life and by the type, energy and abundance of the emitted radiation. Due to its half-life (17 d) and its electron capture decay resulting in the abundant emission of Auger electrons and low energy X rays (20–22 keV), ^{103}Pd can be used for the preparation of seeds which are used as permanent interstitial implants for the treatment of rapidly proliferating tumours. Along with ^{125}I , the radionuclide has been utilized in the treatment of various cancers such as eye, brain, neck, uterus and colon, but at present it is primarily used for prostate tumours. Over the last decade it has proven to be very effective in treating this cancer with minimal side effects.

Palladium-103 is mainly produced by proton bombardment of rhodium ($^{103}\text{Rh}(p,n)^{103}\text{Pd}$). Though soluble rhodium compounds have been tried, at present commercial production involves the use of solid targetry such as powder, wires, foils or electroplated layers. The latter may be prepared by electrodepositing the precious metal on a (copper) target carrier either from

home-made rhodium sulphate or rhodium chloride [8, 9] plating solutions containing appropriate amounts of plating additives (surfactant, depolarizing, stress reducing and/or complexing agents) or from commercially available rhodium plating baths. The plating can be carried out applying DC CVE or CCE and AC CVE (current) electrolysis at elevated temperature (typically 40–60°C). To avoid the purchase of very expensive commercial plating baths, which may be difficult to obtain in developing countries, and to allow reuse of the precious metal target material, an in-house plating/recovery cycle may be preferred.

6.2. TARGET FABRICATION

6.2.1. Target preparation from home-made sulphate plating baths

An in-house plating/electrodissolution/recovery cycle starts with the preparation of a sulphate plating solution from recovered hydrated rhodium oxide. Commercially available sulphate plating baths (Rhodex, Enthone, USA) can also be used to start up.

Recovery of rhodium as hydrated rhodium oxide

Rhodium is recovered as hydrated rhodium oxide from a freshly prepared aqueous RhCl_3 solution obtained by dissolution of the analytical grade compound or from a mixture of hydrochloric acid recovery solutions obtained after centrifugal electro-dissolution of the irradiated rhodium and subsequent extraction of ^{103}Pd .

- (1) Filter the freshly prepared rhodium chloride solution or the combined recovery solutions, containing 2–6 g of rhodium, through an 0.45 μm filter to remove any particles.
- (2) Evaporate the filtrate to near dryness (350°C at the start, 150°C near the end);
- (3) Take up in 350 mL of dematerialized water, heat to 150°C and stir for 15 min.
- (4) Filter through an 0.45 μm filter (2–10 mg of black (Rh) residue may be obtained), cool down the filtrate to room temperature in a water bath.
- (5) Neutralize while vigorously stirring with 10N NaOH to pH = 10–10.5. Use a combined glass electrode as pH sensor.
- (6) To improve the filterability of the yellow $\text{Rh}_2\text{O}_3\text{-aq}$ precipitate, allow digestion for 24 h at 50°C while gently stirring.

- (7) Filter the precipitate on a Bleu Band filter paper (Schleich&Scheull 589) and wash at least ten times with water to remove most of the adsorbed Cl^- anions. As some of the yellow precipitate leaks through the filter, the filtrate is filtered through an $0.45\ \mu\text{m}$ filter.
- (8) Allow both filters carrying the precipitate to dry in the air for 24 h. Remove the rhodium oxide, grind the precipitate in an agate mortar and vacuum dry for 48 h (350 mbar) at room temperature.³

Preparation of a sulphate plating solution from $\text{Rh}_2\text{O}_3\cdot\text{aq}$

To further reduce the Cl^- content of the plating solution, the latter is prepared by dissolution of an appropriate amount of hydrated rhodium oxide in boiling concentrated sulphuric acid followed by dilution with water, addition of a sulphamic acid stress reducing agent and pH adjustment. To simultaneously prepare four targets showing an $11.69\ \text{cm}^2$ surface area and a physical thickness of $40\ \mu\text{m}$, use the following procedure:

- (1) Transfer 4.708 g of hydrated rhodium oxide into a 400 mL beaker provided with a magnetic stirring bar and carefully introduce 10 mL of 95% sulphuric acid. Cover the beaker with a watch glass supported by glass hooks.
- (2) Heat while gently stirring (350°C) until SO_3 fumes evolve and continue for 15 more min. A dark yellow-brown concentrate is obtained.
- (3) Carefully add 250–300 mL of water and continue heating (150°C) and vigorous stirring for 15 min.
- (4) Filter through an $0.45\ \mu\text{m}$ filter and dilute to about 400 mL with water.
- (5) Add 5 g of sulphamic acid and adjust pH to the desired value between 1 and 2 with 10N NaOH.
- (6) Make as much as 450 mL.

Simultaneous plating of four $40\ \mu\text{m}$ Rh target layers (surface area $11.69\ \text{cm}^2$)

Before the rhodium plating, and to ensure good adhesion, the copper target carriers are etched in an acid copper sulphate solution followed by electrodepositing of a $5\ \mu\text{m}$ freshly prepared copper layer in excess. Therefore, two types of plating vessels showing different plating windows are used.

The plating vessels are hollow Perspex cylinders fitted with an axial platinum anode wire mounted in the bottom by means of a tube end fitting with

³ The mean rhodium content of the precipitate equals $49.3 \pm 0.5\%$.

perforated septum. Four symmetrical windows (22.36 or 11.69 cm²) in the vertical wall allow introduction and positioning of four copper target carriers. Each slot is provided with an O ring fitted window, the geometrical shape of which determines the electrodepositing area. Liquid tightness is realized by stainless steel mechanical pestles mounted in a PVC ring surrounding the plating vessel and by pressing the copper carriers against the O ring. The external PVC ring is also fitted with 4 supporting pins to hold a motor stirrer combination in position. The stirrer is a hollow perforated POM cylinder mounted on the axis of the DC motor and surrounding the platinum anode. The stirrer rotation speed is set to 1000 rev./min while the direction of rotation is reversed after 8 s to improve the homogeneity of the deposit.

To maintain the temperature at the required level, a series of six insulated 1 Ω /1 W resistors, through which a suitable DC current is forced (1.1A, 40°C up to 1.8A, 60°C), are circularly mounted at the bottom of the vessel. An insulated sensor is introduced through the stirrer support plate to monitor the bulk temperature of the plating bath. As electrolysis to depletion requires a long time for plating (up to 24 h), evaporation of the plating solution occurs. To maintain the liquid volume at the required value (450 mL), a conductivity glass/graphite sensor monitors the solution level and activates a peristaltic pump supplying distilled water at a rate to compensate for the evaporation losses. The home-made electronics are mounted in a rack and include a motor/stirrer control, an adjustable DC voltage generator card and four V/I converters coupled to four current boosters.

The DC CCE plating to depletion (DR > 98%) is performed at 60°C under vigorous bi-directional stirring (1000 rev./min, 8/8 s) applying a current density of about 6.5 mA/cm².

Use the following plating procedure:

- (1) Clean the copper carriers with abrasive wool, distilled water and acetone; paper dry;
- (2) Mount the carriers in the first Perspex plating vessel (22.36 cm² windows) and add 450 mL of etching/plating solution (250 g CuSO₄.5aq + 25 mL HNO_{3,c} + 50 mL H₂SO_{4,c} in 2.5 L);
- (3) Etch for 3 h while vigorously stirring (1000 rev./min, 8/8 cycle, no current);
- (4) Apply a DC current and plate out copper for 1 h (1000 rev./min, 200 mA/target);
- (5) Empty the plating vessel, remove the carriers, rinse with water/acetone and paper dry;
- (6) Mount the pretreated copper carriers in the rhodium plating vessel (11.69 cm² windows);

- (7) Introduce the rhodium plating solution preheated to 60°C;
- (8) Apply current (75 mA/target), stir (1000 rev./min, 8/8 cycle) and switch on the power supply of the heating element (1.8 A) and that of the water feedback system;
- (9) Continue plating for 24 h (Rh depletion \geq 98%);
- (10) Empty the plating vessel, remove the targets, rinse them with water/acetone and paper dry;
- (11) Remove any superficial Rh black with a scouring suspension, rinse with water and acetone and air dry (rhodium loss of as much as 2 mg).

6.2.2. Target preparation from home-made chloride plating baths

Rhodium chloride plating solutions can be prepared by dissolution of the analytical grade compound or from rhodium recovery solutions obtained after electrosolubilization of irradiated targets and extraction of palladium. Commercially available plating solutions (Degussa, Germany) containing some plating additives can be used as well. Direct current CCE or AC CVE plating to a depletion level larger than 98% at about 40°C can be applied.

Direct current CCE plating at 40°C

The following procedure can be used for the simultaneous preparation of four targets with a surface area of 11.69 cm² and a physical thickness of 48 μm. The preliminary treatment of the copper target carriers and the plating set-up are described below.

- (1) Dissolve an amount of hydrated RhCl₃ containing 2.8 g of rhodium in 400 mL of water. Alternatively, the filtered (0.45 μm filter) combined recovery solutions containing the same weight of rhodium can be evaporated to near dryness (350°C at the start, 150°C near the end) and residue taken up (gentle stirring, 50°C) in 400 mL of distilled water.
- (2) Neutralize the solution with 10N NaOH up to pH2–2.5. Use a combined glass electrode as sensor.
- (3) Filter the solution through an 0.45 μm filter and adjust the volume to 450 mL.
- (4) Heat to 40°C then transfer the solution to the plating vessel.
- (5) Apply a DC CCE of 100 mA per target, set the heating current to 1.1 A and switch on the water feedback system.
- (6) Continue plating for 24 h (Rh depletion \geq 98%).
- (7) Empty the plating vessel, remove the targets, rinse them with water/acetone and paper dry.

- (8) Remove any superficial Rh black with a scouring suspension, rinse, and dry as in point (7). The surface cleaning results in a rhodium loss of as much as 2 mg.

Alternating current CVE plating at 45°C

A single target can be AC plated (11.69 cm² surface area, rhodium layer thickness 40 μm) using a rhodium chloride/ammonium chloride bath in a rectangular perspex plating vessel (14 cm × 14 cm × 30 cm) fitted with a single plating window in the bottom and a rectangular platinum foil counter electrode (8 cm × 1 cm) mounted parallel with the target carrier at a distance of 1 cm.

- (1) Dissolve 50 g of RhCl₃.6aq and 5 g analytical grade NH₄Cl in 500 mL of distilled water;
- (2) Clean the target carrier with acetone and water and mount it in the plating window;
- (3) Heat up the plating solution to 45°C, pour 300 mL into the plating vessel and start gentle heating (45°C);
- (4) Homogenize the bath by bubbling nitrogen;
- (5) Apply an asymmetric square wave (total time 20 s, cathodic wave 15 s, anodic wave 5 s) plating voltage for 2.5 h;
- (6) Empty the plating vessel, remove the target, rinse with water and acetone and dry.

6.2.3. Other solid rhodium targets

As other solid target materials, rhodium foils (10–100 μm), rhodium wires (0.5 mm) or rhodium powder can be used. In either case, centrifugal electrodisolution combined with wet chemistry can be used for the separation of ¹⁰³Pd. Foils, wires and fragmented rhodium allow partial recovery of ¹⁰³Pd when the thermal diffusion technique is applied.

6.3. IRRADIATION OF ELECTROPLATED RHODIUM TARGETS

To take full advantage of the ¹⁰³Rh(*p,n*)¹⁰³Pd excitation curve and avoid the formation of the radionuclidic ¹⁰¹Pd impurity (¹⁰³Rh(*p,3n*)¹⁰¹Pd) the proton entrance energy should be less than 20 MeV (typically 18 MeV, between 18 and 20 MeV the ¹⁰³Pd yield increases by less than 4%). With an 18 MeV entrance energy only trace amounts of long lived ^{102m,102g}Rh are formed. Though efficient radiochemical processing will prevent radionuclidic contamination of the ¹⁰³Pd

bulk with these nuclides, they will accumulate when the target material is recycled. Therefore, appropriate radiation protection measures should be taken in recycling procedures.

The physical thickness of the rhodium layer should be such that for a given beam/target angle geometry the particle exit energy should be 6 MeV. For 90° geometry, this means 475 μm. To minimize the physical thickness of the rhodium layer (and hence the cost per target), a 6° geometry is preferable, in which case a 45–50 μm layer is recommended. If high beam current irradiations (>>200 μA) are intended, the thickness of the copper backing should be reduced to 1 mm (tolerating a 30 bar coolant pressure), the coolant flow rate should be as high as possible ($v_{\text{coolant}} = 5000 \text{ cm/s}$) and circular beam wobbling should be tried.

6.4. TARGET PROCESSING

If the aim is to produce a ^{103}Pd solution suitable for seed production by electrochemical techniques (e.g. electrolysis, exchange reactions), solubilization of the target material followed by a wet chemistry ^{103}Pd separation or a non-destructive thermodiffusion extraction of the radionuclide is necessary. In general, the solubilization is the most difficult production step.

Irradiated rhodium metal targets (plated layers, foils and wires) have frequently been dissolved by sodium bisulphate fusion (time consuming, complex medium), by gold tetrachloroaurate oxidation [10] (very expensive, time consuming) and by static AC electrodisolution in hydrochloric acid [11].

Up to now the latter method has been recommended for the solubilization of foils, but is not applicable for rhodium powder, wires or fragments. Moreover, it suffers from a number of limitations such as incomplete dissolution due to partial fragmentation, limited and non-time controlled solubilization, and large volumes of hydrochloric acid required for gram amounts of target material. A new time controlled centrifugal high current density electrodisolution technique resulting in quantitative (>99%) solubilization of as much as 3 g of target material in a limited volume (40 mL) of acid has been developed.

6.4.1. Centrifugal electrodisolution technology

Upon removal of the copper backing of an irradiated target in a flow through stripper using concentrated nitric acid and plenty of rinsing water, the rhodium is recovered as crude fragments which are mechanically ground into

finely divided fragments. The latter are introduced into a centrifugal electro-dissolution unit allowing time controlled dissolution of the fragments up to a required dissolution level (99%).

The electro-dissolution system consists of a hollow cylindrical electrographite working electrode (WE) receptacle fitted with an insulating polyetheretherketon disk at the bottom and a hollow cylindrical PERSPEX upper part to increase the volume of the system. An electrographite CE rod extending to one mm of the PEEK disk is mounted on the axis of the receptacle. The latter is mounted on the axis of a DC-motor/tacho combination operating at 1000 rev./min. Connecting an AC power supply to two parallel circuited disposable brass brushes which are in contact with the axis of the motor and the graphite CE, a maximum of 30 A AC current may be forced through the concentrated hydrochloric acid solution containing the rhodium fragments inside the receptacle. Under the influence of centrifugal force and heat generated fluid convection currents, the rhodium fragments are collected in the lower section of the vertical inner wall of the WE, where the concentration of the electroactive chlorine species dissolving the fragments is maximized. Appropriate feedback systems allow temperature (80°C), volume (40 mL HCl) and current (to 30 A, typically 25 A) to be kept constant. The optimum input and feedback concentration of HCl is 12N. The final concentration is 7.3N HCl.

The dissolution rate dW/dt (mg Rh dissolved per minute) can be represented by a transcendental mathematical expression (Eq. 12):

$$dW(t)/dt = K \cdot \{1 - \exp(-b \cdot C(t))\} \cdot \{1 - \exp(-c \cdot \Delta I)\} \cdot \{1 - \exp(d \cdot W(t))\} \quad (12)$$

$$C(t) = [C_{eq} \cdot \{1 - \exp(-a \cdot t)\}] - C_0 \cdot \exp(-a \cdot t) \quad (13)$$

$$C_{eq} = C_0 \cdot (1 - e \cdot I) \quad (14)$$

with $C(t)$ equal to the bulk concentration of HCl as a function of time and C_{eq} the equilibrium concentration at $t = \infty$. In the equations a , b , c , d and e are experimental constants, I the applied current, C_0 input and feedback concentration of HCl. $\Delta I = I - I_{co}$ can be considered to be the effective applied current with I_{co} the cut-off current below which the dissolution rate is reduced to a negligible level.

A numeric integration algorithm allows calculation of the dissolution rate, of the amount of dissolved rhodium as a function of time and of the time (T) required to dissolve the input weight of rhodium to an acceptable solubilization level (99%), $T(99\%)$. The single run capacity of the present system amounts to 3 g of rhodium while the technology can be used to dissolve

not only fragments obtained from electroplated targets, but also for the solubilization of irradiated rhodium powder, cut wires and cut foils as can be seen from Table 3.

Table 4 shows that the reproducibility of the system is excellent. The table applies to electroplated targets with a surface area of 11.7 cm² and a physical thickness of 55 μm. When input and feedback concentration of HCl equals 12N, the final concentration of the acid is 7.3N ± 0.3N.

TABLE 3. APPLICABILITY OF THE CENTRIFUGAL ELECTRO-DISSOLUTION TECHNOLOGY (T(99%) = CALCULATED TIME TO DISSOLVE 99% OF RHODIUM)

Type of Rh	Weight of Rh (g)	Rh dissolved (%)	Time (99%) (min)
Fragments	0.8070	98.7	141
Powder	2.5467	99.8	167
Powder	2.5053	99.6	167
Cut wire	1.1269	99.7	145
Cut foil	1.3340	99.8	149

TABLE 4. REPRODUCIBILITY OF THE CENTRIFUGAL ELECTRODISSOLUTION SYSTEM (M ± SD = MEAN ± SD)

Number	Input Rh (g)	Solubilized Rh (g)	Dissolved (%)	Volume (mL)	Final HCl normality
1	0.8070	0.7965	98.7	42	7.38
2	0.8046	0.7942	98.7	40	7.36
3	0.8083	0.8043	99.5	41	7.48
4	0.8080	0.8033	99.4	39	7.40
5	0.8054	0.7975	99.0	38	7.20
6	0.8076	0.8036	99.5	39	7.20
7	0.8088	0.8025	99.2	40	7.20
8	0.8058	0.7986	99.1	39	7.32
9	0.8042	0.7998	99.5	40	8.0
10	0.8024	0.7948	99.0	38	7.0
M ± SD	0.8060 ± 0.0021	0.8000 ± 0.0038	99.2 ± 0.3	39.6 ± 1.3	7.3 ± 0.3

6.4.2. Separation of carrier free ^{103}Pd

Wet chemistry methods

Palladium-103 is mostly separated from the solubilized rhodium matrix by solvent/solvent extraction, anion or cation exchange chromatography or electroseparation, i.e. separation by CCPE.

For solvent extraction the 3N HCl/furyldioxime-chloroform system [12] should be applied as it does not require evaporation of the solution obtained after centrifugal electrodisolution. Only a twofold dilution with water before the extraction is necessary. The application of ion exchange chromatography requires foregoing evaporation to dryness. For cation exchange chromatography the residue is taken up in 0.1–0.4N HCl. Rhodium and palladium are then separated on an AG50WX8 (H^+ /100–200 mesh grade) column (1.5 g resin). Rhodium is eluted first, after which palladium is collected in 10 mL HCl_c .

Anion exchange involves the dissolution of the residue in 0.03N HCl and the Cu/Rh/Pd is separated using a Dowex1X8(Cl^- /100–200 mesh grade) column (1.5 cm \times 10 cm). Copper is eluted with 0.03M HCl, rhodium with 6M HCl and palladium with a 1:1 mixture of 0.5M $\text{NH}_3/\text{NH}_4\text{Cl}$. To apply electroseparation, the 7.3N HCl solution should be partially neutralized (1–2M HCl). The rhodium is then plated out on a cathode, the potential of which is set at -0.54 V versus an $\text{Ag}/\text{AgCl}_{\text{sat}}$ reference electrode. The disadvantage of this method is that the rhodium target material cannot be recycled in a simple way.

Thermal diffusion

Thermal diffusion involves the migration of the carrier free ^{103}Pd to the surface area of foils, fragments or wires under the influence of heating up (in the air or in a vacuum) to a temperature higher than 1000°C . Upon cooling the palladium can be dissolved in a suitable acid, typically HCl. Though extremely simple, one main disadvantage is the limited extraction yield. This is illustrated in Table 5. The surface area of the foils in both experiments was 1 cm^2 . Obviously, the extraction yield depends on the chemical purity of the rhodium and on the temperature. As the melting point of rhodium metal is 1966°C , higher temperature (as much as 1900°C) diffusion should be tried.

TABLE 5. THERMAL DIFFUSION/EXTRACTION OF ^{103}Pd FROM RHODIUM FOILS

Foil thickness (μm)	Beam/target angle ($^\circ$)	Irradiation beam	Irradiation time (h)	Thermal diffusion	Pd yield (%)
2 × 25	90	18 MeV 30 μA	2	1400°C, vacuum 30 min	70
50	7	29 MeV 20 μA	1	1200°C, air 2 h	50

6.5. QUALITY CONTROL

6.5.1. Target quality control

Three criteria are used to estimate the quality of electroplated rhodium targets, namely the homogeneity of the rhodium layer, the SAG and its behaviour during a TST.

The homogeneity of the rhodium layer is important because it may strongly affect the local production rate of ^{103}Pd . The homogeneity is easily determined by micrometry. Taking at least 12 measuring points distributed over the whole surface area, the SD of the data set should be less than or equal to 5%.

The granulometry can be evaluated by comparing the X250 magnification SEM shots of a given target with those of an ‘accepted’ reference. The latter may be a rhodium target plated from a commercially available rhodium plating solution (RHODEX, Enthone, USA). Visual inspection may result in a qualification such as ‘comparable’. The TST involves heating of the target to 500°C (= maximum allowable temperature of the rhodium layer during the irradiation) for one hour, followed by the submersion of the hot target in cold (15°C) water and by multiple bending of the target carrier. No crack formation or peeling off of the rhodium layer should be observed.

The TST is a measure of the incorporation of plating bath compounds into the rhodium crystal lattice and/or of their presence at the carrier/rhodium layer interface. In both cases, irradiation will result in loss of target material, peeling off, and a decrease in the yield of ^{103}Pd .

The targets prepared from home-made sulphate baths (Section 6.2.1) and from commercially available plating solutions (Rhodex, Enthone, USA) satisfy all three criteria. Targets obtained from home-made chloride solutions are

homogeneous but do show a poor granulometry compared with that of the reference, and withstand a temperature shock test only up to 200°C. Moreover, upon dissolution of the copper backing, the rhodium layer disintegrates into a black powder (not into silvery white fragments as do the Rhodex reference and the targets prepared from home-made sulphate baths), the dissolution kinetics of which do not satisfy Eq. (12). These targets should be used only for the production of limited quantities of ^{103}Pd .

6.5.2. Radiochemical quality control

The final radiochemical used for seed preparation should preferably be a carrier free (specific activity $\gg 50$ Ci/g) palladium chloride solution in dilute ammonium hydroxide ($7 < \text{pH} < 10$). Commercially available solutions show an activity concentration higher than 500 mCi/mL. The accepted radionuclidic purity is $\geq 99.95\%$ ^{103}Pd .

6.6. CONCLUSIONS OF SECTION 6

Batch production of ^{103}Pd solutions suitable for the preparation of brachytherapy seeds by electrolysis or spontaneous exchange generally involves the 18 MeV proton irradiation of rhodium metal followed by dissolution of the target material and separation of the carrier free palladium.

Powder, wires, foils and electroplated rhodium layers can be used as solid material. In either case the effective thickness of the target material should be 40–50 μm when the beam/target angle equals 6° . Minor quantities of longer lived rhodium isotopes ($^{102\text{m}}, ^{102\text{g}}\text{Rh}$) formed should be taken into account when the expensive target material is recycled.

Electroplated rhodium targets can be prepared from home-made or commercially available sulphate or chloride baths containing appropriate plating additives. When home-made plating solutions are used, the addition of a 1% sulphamic acid stress reducing agent is recommended. As a plating technique, CCE DC or AC electrolysis at elevated temperature (40–60°C) can be applied. As the plating CE is less than 100%, plating to depletion ($>98\%$ rhodium deposition) is recommended. For high current beam irradiations, targets prepared from sulphate baths are preferable as they satisfy the homogeneity (5%), the granulometry (comparable with Rhodex reference) and the TST (500°C) criteria.

For the dissolution of the irradiated solid target material, time controlled centrifugal electrodisolution at 85°C and 2 A/cm⁻² WE current density are preferable to techniques such as bisulphate fusion (time consuming, complex

medium), gold tetrachloroaurate oxidation (very expensive) or static AC electrodisolution in 6N hydrochloric acid (non-quantitative, limited to foils). When 12N hydrochloric acid is used as input and feedback concentration, it allows the solubilization of as much as 3 g of powder, wires, foils and fragments in 40 mL within less than 3 h.

Carrier free ^{103}Pd can be separated from the dissolved rhodium matrix by solvent/solvent extraction and ion exchange chromatography. When centrifugal electrodisolution is used, solvent extraction with the HCl/furyldioxime-chloroform system is preferable, as it involves no prior time consuming evaporation. If ion exchange chromatography is involved, cation exchange should be recommended. Though attractive, the electroseparation suffers from one main disadvantage, namely that the target material cannot be reused simply. The plating/electrodisolution/recovery cycle illustrates the reuse of the precious and expensive target material.

7. RESULTS OF THE WORK ON TARGET FABRICATION FOR ^{201}Tl PRODUCTION

7.1. INTRODUCTION

Cyclotron produced radionuclides are used routinely for the diagnosis of an extensive variety of diseases. Contrary to expectations, ^{201}Tl continues to be one of the most broadly used SPECT radioisotopes as a myocardial perfusion imaging agent. In addition, ^{201}Tl has recently been tried as a diagnostic agent for tumour detection [13].

Thallium-201 is routinely produced by proton bombardment of solid electroplated ^{203}Tl (>>98%) targets by the reaction $^{203}\text{Tl}(p,3n)^{201}\text{Pb}$ followed by decay of the ^{201}Pb into ^{201}Tl . For the most part, until a few years ago the target material was electroplated on a copper carrier from sulphuric or perchloric acid TlNO_3 , TlClO_4 or $\text{Tl}_2(\text{SO}_4)_2$ solutions containing organic quality improving agents such as aromatic amines (o-toluidine), phenols (o-cresol) and polypeptides (peptones) [15].

Application of DC CVE electroplating resulted in competitive hydrogen evolution and in the incorporation of the plating additives into the thallium metal lattice or at the metal/carrier interface. The deposits showed very poor surface area quality (post-plating mechanical pressing was used to smooth the surface) and poor homogeneity, and could withstand only a limited beam

current intensity. Frequently, thermal destruction of the incorporated organic material during bombardment gave rise to crater formation and peeling off of the layer, resulting in losses of enriched material and poor reproducibility of the $^{201}\text{Pb}/^{201}\text{Tl}$ production yield.

Recently, an inadequate supply of ^{201}Tl due to the limited capability of the solid ^{203}Tl targets to withstand high beam currents ($\gg 200\ \mu\text{A}$) became a major obstacle in meeting the ^{201}Tl needs of nuclear medicine. The poor target quality problem may be avoided by applying a new AC CCE plating technology involving the use of an alkaline plating bath containing appropriate amounts of a non-incorporated complexing agent, of a surfactant and of an anodic depolarizer. Simultaneously a fast and quantitative electrochemical technique for the recovery of the expensive enriched ^{203}Tl target material from irradiated and processed targets was developed

7.2. TARGET FABRICATION

The plating/recovery cycle starts with preparation of an 0.5M alkaline EDTA (complexing agent) solution ($\text{pH} > 12.5$) containing 1% hydrazine hydrate as an anodic depolarizer (avoids oxygen generation at the electrodes) and 0.2% BRIJ-35 as tensio-active agent (smooths the surface). Commercially available $^{203}\text{Tl}_2\text{O}_3$ or electrochemically recovered $^{203}\text{Tl}_2\text{SO}_4$ can be used as ^{203}Tl compounds. To avoid concentration polarization (that enhances the risk of dendrite formation) the ^{203}Tl content of the plating bath is such that only a limited fraction (DR) of the metal is required for target plating. The plating current density is adjusted so that the CE (ϵ) of the diffusion controlled plating process is higher than 99%, allowing time controlled layer thickness preparation.

The relationship between the maximum allowable current density (i_A) and the required ^{203}Tl concentration at the end of a plating batch (C_c) was determined experimentally. This relationship allows calculation of the optimum thallium concentration at the start of the electrolysis (C_0) to ensure $\epsilon > 99\%$ for a given current density and target layer geometry (surface area, thickness, number of targets plated per run).

7.2.1. Preparation of the alkaline EDTA plating solution

The following alkaline ^{203}Tl containing EDTA solution, holding appropriate amounts of BRIJ-35 surfactant and hydrazine depolarizer, is used for the repetitive preparation of four external beam thallium targets (surface area $11.69\ \text{cm}^2$, thallium layer thickness $78\ \mu\text{m}$, Tl weight $1092\ \text{mg}$) used in a 6°

TABLE 6. CHEMICALS FOR ALKALINE EDTA PLATING TECHNOLOGY

Tl ₂ O ₃	Enriched ²⁰³ Tl oxide, compound obtained by the purchase of the enriched material
²⁰³ TIEDTA ³⁻	Final product obtained in the VUB recovery cycle of the enriched material
EDTA	(ethylene-dinitrilo-tetra-acetic acid, di-sodium salt) — Titriplex III p.a. (Merck-1.08418)
NaOH	Sodium hydroxide pellets p.a. (Merck-6495)
Hydrazine hydrate	(55%) (Aldrich-22.581-9)
BRIJ-35	(30%) (Technicon Diagnostics-T 21-0110-17)
H ₂ SO ₄	(95–97%) p.a. (Merck-731)
Perhydrol	(30%) p.a. (Merck-1.07209)
H ₂ O	Demineralized water — conductivity < 0.1 μS/cm
Acetone	(purum) (Belgolabo 94-90013)
Parafilm "M"	American National Can
PH indicator strips	(Merck 9535)
Abrasive wool	Type 1: F12/002 very fine (abrasive centre) Type 2: F12/012 ultra fine (abrasive centre)

beam/target irradiation geometry and irradiated with 28.5 MeV proton beams. The target layers are deposited on copper carriers. The chemicals used are summarized in Table 6.

Though the volume of the plating bath is not very critical, it should be kept between 420 and 450 mL. The procedure is as follows:

- (1) Put 360 mL demineralized water in a 600 mL beaker (B1) fitted with a Teflon coated magnetic stirring bar.
- (2) Add 84 g of solid EDTA and 20 g of NaOH pellets and stir until a colourless solution is obtained.
- (3) Add 10 mL hydrazine hydrate and 1 mL BRIJ-35 and homogenize (solution 1).
- (4) Transfer 12.20 g of enriched ²⁰³Tl₂O₃ to a second 600 mL beaker (B2) also fitted with a magnetic stirrer bar. Alternatively a suitable volume of recovery solution containing the same amount of thallium can be used.
- (5) Transfer solution 1 into beaker B2 by means of a peristaltic pump at a flow rate of 10 mL/min. Cover beaker B1 with Parafilm.

- (6) Reduce the stirring rate of the stirring bar in B2 and rinse the wall of this beaker with 1 mL hydrazine hydrate (if any yellow-brown colour persists in the foam layer, gently swirl the beaker until the colour disappears). Check pH by means of an indicator strip. The pH should be >12.5.
- (7) Continue slow stirring of B2 for five more minutes.

Solution 2 (in B2) is now ready for electrodeposition. The plating solution can be used as many as ten times. After each batch the solution should be replenished with an appropriate amount of thallium.

7.2.2. Simultaneous plating of four 78 μm thallium target layers (surface area 11.69 cm^2)

The plating vessel is a hollow Perspex cylinder fitted with an axial platinum anode wire mounted in the bottom by means of a tube end fitting with a perforated septum. Four symmetrical windows (11.69 cm^2) in the vertical wall allow introduction and positioning of four copper target carriers. Each slot is provided with a window fitted with an O ring, the geometrical shape of which determines the electrodepositing area. Liquid tightness is realized by stainless steel mechanical pestles mounted in a PVC ring surrounding the plating vessel and by pressing the copper carriers against the O ring. The external PVC ring is also fitted with four supporting pins to hold a motor stirrer combination in position. The stirrer is a hollow perforated POM cylinder mounted on the axis of the DC motor and surrounding the platinum anode. The stirrer rotation speed is set at 1000 rev./min while the direction of rotation is reversed after 8 s to improve the homogeneity of the deposit. The plating is carried out at room temperature. Electrolysis at higher temperature (60°C) results in a reduction of thallium in the bulk of the solution by hydrazine.

The AC generator plating waveform is an asymmetric bi-polar ($I_c/I_a = 3/2$) chopped ($f_{\text{ch}} = 1000$ Hz) sawtooth ($f_{\text{st}} = 100$ Hz) with a duty cycle of 60% ($t_c/t_a = 6/4$). The generator output is fed into four independent V/I converters — current boosters that force a net and constant cathodic current through the plating solution. As the plating CE is higher than 99%, time controlled plating can be performed. The optimum current density is 2.14 mA/cm^2 . The current density may be increased if the DR is decreased accordingly.

Before the plating, the carriers are cleaned in the following way:

- (1) Rub the deposition area of the copper backing successively with abrasive wool type 1 and type 2 for one minute, respectively;
- (2) Rinse with water and acetone;
- (3) Dry the carriers with adsorbing paper and weigh;

- (4) Introduce the backings in the four slots of the deposition vessel and turn the knobs of the mechanical pestles clockwise in pairs to ensure liquid tightness.

Electrodepositing of four targets is carried out after the transfer of solution 2 to the plating vessel fitted with four cleaned target carriers:

- (1) Pour solution 2 into the plating vessel. Do not rinse the beaker but cover it with Parafilm.
- (2) Connect the 4 outputs of the current boosters to the cathodes.
- (3) Introduce with care the electromagnetic stirrer so that the central platinum anode wire fits into the hollow perforated stirring cylinder.
- (4) Start stirring motor (rotational speed: 1000 rev./min, 16 s, 8/8 s).
- (5) Start the electrolysis by connecting the anode to the ground and note the time. The four current indicators should give a 25.0 ± 0.2 mA reading after a few minutes of deposition. With a 100% CE a ^{203}Tl deposition rate of 7.57 mg.mAh^{-1} is obtained, for a $1092 \text{ mg } ^{203}\text{Tl}$ deposit per target the plating time equals 5.77 h, i.e. 5 h 46 min.
- (6) After 5 h 46 min of electrolysis, stop the stirring motor and carefully remove the stirring assembly. Do not rinse the perforated hollow stirrer cylinder, but quickly introduce it into a 250 mL Perspex holder.
- (7) Pour the 40% depleted plating solution into beaker B2. Rinse the deposition vessel with 3 times 15 mL of demi-water and collect the rinsing liquid in a third 250 mL beaker (B3). Cover beaker B3 with Parafilm.
- (8) Remove and rinse each of the targets extensively with hard water (waste), followed by rinsing with demi-water (waste).
- (9) Dry the targets by means of adsorbing paper and weigh. Determine the CE by calculating:

:

$$\eta_i = \frac{W_i}{1092} * 100 \quad (15)$$

where:

η_i = CE for target I

W_i = Tl weight (mg) of target i

The CE should be $100 \pm 0.2\%$.

- (10) Wipe off the surface oxide layer with adsorbing paper and store the targets in a vacuum desiccator or under nitrogen. Wiping off may result in a loss of a few milligrams of ^{203}Tl . If not acceptable, the wipe test samples should be collected and later destroyed in an $\text{HClO}_4/\text{H}_2\text{O}_2$ mixture. After H_2O_2 has been catalytically eliminated by introducing a platinum electrode, thallium can be bulk recovered in this solution.

7.2.3. Preparation of the second (to the tenth) plating bath

As stated previously, the partially depleted bath can be reused for plating after the addition of a suitable amount of thallium:

- (1) Transfer 4885 mg enriched $^{203}\text{Tl}_2\text{O}_3$ to beaker B1 fitted with a magnetic stirring bar.
- (2) Transfer the 40% depleted plating solution from beaker B2 to B1 by means of a peristaltic pump at a flow rate of 10 mL/min. Cover beaker B2 with Parafilm.
- (3) Reduce the stirring rate in B1 and rinse the walls of this beaker with 1 mL hydrazine hydrate (if any yellow-brown colour persists in the foam layer, gently swirl the beaker until the colour disappears).
- (4) Continue slow stirring of B1 for five more min. Check pH by means of an indicator strip. The pH should be >12 , if not add a few drops of 10% NaOH.

These procedures can be repeated as many as 10 times, whereupon due to degradation of the surfactant bulk recovery of the 40% depleted bath is necessary. This should be done by CCPE.

7.2.4. Optimum current density range and C_c

The foregoing procedure applies to the simultaneous production of ^{203}Tl targets with a surface area of 11.69 cm^2 that are to be irradiated in a 6° beam/target geometry with a 1 cm diameter collimator. The optimum plating current density thus equals 2.14 mA/cm^2 and the 40% DR of the plating bath corresponds to a thallium concentration of 14.3 g/L at the end of a plating batch. This concentration is the C_c , i.e. the minimum concentration of thallium at the end of the electrodeposition required to ensure an overall plating CE for a given current density of $\geq 99\%$. This means that for given current density and thallium concentrations $C \geq C_c$, the diffusion controlled thallium supply rate from the bulk to the solution/cathode interface is equal to or higher than the

plating deposition rate governed by Fick's law and, as a consequence, that concentration polarization is excluded.

For the plating set-up described above, the C_c in g/L as a function of the applied current density (i_A in mA/cm²) was determined experimentally and may be calculated from Eq. (16) that holds from 0.5 mA/cm² to 10 mA/cm²:

$$C_c = 0.822 * i_A + 12.80 \quad (16)$$

Equation (16) can be used to calculate the required input concentration C_0 (g/L) of thallium at the start of the plating as a function of i_A , the plating surface area per target S (cm²), the plating time t (h), the volume V (L) of the plating bath and the number n of targets plated per batch ($n = 2, 4$ or 8):

$$C_0 = [0.07 + (0.00757 * t * n / V)] * S * i_A + 12.82 \quad (17)$$

Equation (17) takes into account that the maximum deposition rate of monovalent 98% enriched ²⁰³Tl is equal to 0.00757 g·mA/h (100% plating CE).

Equations (16, 17) allow evaluation of the optimum current density and plating bath composition as a function of the target layer and irradiation geometry. Up to now the technology has been applied to produce targets for eight ²⁰¹Tl production facilities.

7.2.5. Recovery of enriched thallium

The enriched thallium from partially depleted plating solutions (alkaline EDTA solutions containing thallium metal only) or from recovery solutions (nitric or sulphuric acid or alkaline ammonia nitrate solutions containing Cu, Tl and carrier free ⁶⁵Zn) can be recovered by CCPE after processing of irradiated targets. This electroseparation technique is based on the shift of a metal ion discharge interval and subsequent selective separation by addition of complexing agents to and/or pH adjustment of a solution containing a mixture of metals. The method involves the use of a three electrode set-up:

- (1) A large area cylindrical copper (or platinum) gauze cathode;
- (2) A platinum wire anode mounted axially inside the cathode;
- (3) A reference electrode positioned close to the cathode.

When immersed in a recovery solution, the composition of which is properly adjusted, the electrode set is connected to a potentiostat. This allows fixing of the potential of the cathode versus the reference electrode to a value at which one metal is selectively and quantitatively deposited. Upon removal of

TABLE 7. ELECTROSEPARATION OF COPPER, THALLIUM AND ZINC. REFERENCE ELECTRODE Hg/HgSO₄/sat K₂SO₄ (+412 mV VERSUS NHE)

Element	Solution	pH	Cathode	E _{cathode} versus Ref
Cu	HNO ₃ /H ₂ SO ₄	1.5 < pH < 3	Cu gauze	-0.630
Tl	EDTA	>12	Pt gauze	-1.650
Zn	NaOH excess	>>12	Cu gauze	Constant current

the metal deposit from the cathode, or upon introduction of a new cathode, the composition of the solution and the potential of the cathode are changed so that a second metal can be plated out and so on. The end of deposition of a given metal is detected by monitoring the exponential current flowing through the cathode circuitry, current that should approach zero near the endpoint.

The experimental conditions for the separation of copper, thallium and zinc are summarized in Table 7. Up to 15 g of enriched ²⁰³Tl may be recovered in a single run.

Separation of copper

Copper may be present in the recovery solutions if nitric acid is used to dissolve the thallium layer. If dissolution of the irradiated target material is performed in sulphuric acid, this step may be skipped. This is also the case when thallium is recovered from partially depleted alkaline EDTA solutions. So that this method can also be applied when ²⁰³Tl/²⁰¹Pb is separated by coprecipitation of carrier free ²⁰¹Pb with Fe(OH)₃ in alkaline ammonia nitrate medium, the procedure starts with the addition of concentrated ammonia to nitric acid recovery solution.

Use the following procedure:

- (1) Add concentrated ammonia to the recovery solution containing as much as 15 g Tl in 300 mL, until a deep blue colour appears.
- (2) Neutralize the solution with concentrated H₂SO₄ until the colour changes from deep blue to light blue-green. Add 1 mL H₂SO₄ in excess and 1 mL N₂H₄ (51%). Check the pH of the solution; it should be between 1.5 and 3.
- (3) Introduce a large area copper gauze/platinum wire electrode set and electrolyze for 2 h by CCPE with E_{cathode} = -0.630V versus mercury/mercury sulphate reference electrode.

- (4) Remove the electrode set from the solution and rinse it with water that is collected in the recovery solution.

The copper gauze electrode will be reused to deposit the ^{65}Zn after the addition of a suitable amount of zinc carrier.

Separation of thallium

Upon removal of copper, the composition of the recovery solution is altered so that it approaches that of the plating bath described in Section 7.2.1. Thallium-203 is selectively plated out on a large area platinum gauze electrode, from which it can be re-dissolved in diluted sulphuric acid containing a small amount of hydrogen peroxide to increase the dissolution rate. The solution thus obtained can be used to simply prepare a new plating bath in the following way:

- (1) Add 50 mL NaOH 10N, 10 g solid EDTA, 1 mg ZnSO_4 hold back carrier and 1 mL 55% N_2H_4 hydrate depolarizer to the solution.
- (2) Introduce the platinum gauze/platinum wire electrode set and electrolyze for 2 h at $E_{\text{cathode}} = -1.650$ V versus mercury/mercury sulphate reference electrode.
- (3) Remove the electrode set from the solution and introduce it in an aqueous solution (pH = 12) for 30 min, keeping the electrolysis voltage switched on (= rinsing the thallium deposit).
- (4) Remove the electrode set from the rinsing solution and dissolve the deposited Tl in 100 mL H_2O containing 1 mL $\text{H}_2\text{SO}_{4,c}$ and 1 mL $\text{H}_2\text{O}_{2,c}$ while stirring and gently heating (50–60°C). Leave the platinum gauze electrode in the solution and boil for 15 more min to catalytically destroy the excess H_2O_2 .
- (5) A part of the resulting solution, showing a thallium content of 10.92 g, may be directly used for preparation of a new plating solution. Simple addition of EDTA (84g), NaOH (20g), N_2H_4 (10 mL) and BRIJ (1 mL) and make up to volume (450 mL) allows target plating as described in Section 7.2.2.

Separation of zinc

Though of no practical concern for radionuclide production itself and after the addition of a suitable amount of carrier, ^{65}Zn is plated out on the copper gauze electrode used solely to avoid the creation of large volumes of long lived liquid waste, the disposal of which is very expensive. Pressing together the copper gauze after the zinc plating, less than 1 cm³ of solid waste is

obtained. As the precious enriched ^{203}Tl has been removed in the preceding step, CCE electroplating at high current density can be applied.

- (1) Add 100 g NaOH and 4.5 g $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ and stir until a clear solution is obtained. Then add 50 mL N_2H_4 (51%);
- (2) Introduce the copper gauze/platinum wire electrode set and electrolyze at CCE for 45 min ($I = 20 \text{ A}$);
- (3) Add 25 mL N_2H_4 (51%) and 4 g $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ (dissolved in 20 mL NaOH 10N and 30 mL H_2O) and continue plating for up to two hours;
- (4) Remove the electrode set from the solution, rinse with water and air dry.

The resulting zinc coated copper gauze can be compressed and disposed of as solid waste.

7.3. IRRADIATION OF ELECTROPLATED THALLIUM TARGETS

To limit the radionuclidic contamination of the ^{201}Tl bulk with ^{200}Tl (half-life 26.1 h) formed by the nuclear reaction $^{203}\text{Tl} (p,4n)^{200}\text{Pb} \rightarrow ^{200}\text{Tl}$ to an acceptable level, the entrance energy of the impinging particles should be set at 28.5 MeV. This means that the practical yield is only half the maximum value that can be obtained by irradiation with 36 MeV protons. With a physical thickness of the thallium layer equal to 780 μm irradiated in a 0° beam/target geometry, the exit energy is about 19.4 MeV, thus taking full benefit of the lower energy part of the excitation function of the $(p,3n)$ reaction.

As the thermal conductivity (0.461 W/cm/K) of thallium metal is poor and the melting point (303.5°C) low, the power density distribution on target, and hence the risk of melting the target material, can be reduced by limiting the physical thickness of the layer to 78 μm and irradiation under a 6° angle. To avoid local melting, circular wobbling is strongly recommended, in which case the collimator design should take into account the associated current burden.

With a 2 mm thick flat copper carrier and a coolant channel having a rectangular cross-section (length 1.2 cm; width 0.25 cm; cross-sectional area 0.3 cm^2), the targets described previously can withstand a beam current of as much as 300 μA when a high coolant flow rate (50 L/min) and appropriate circular wobbling (wobbler radius $R_w = 3.37 \text{ mm}$) are applied. The radius of the cylindrical collimator equals 5 mm, the corresponding collimator current burden is limited to about 80 μA and the target current density is 0.5 $\mu\text{A}/\text{cm}^2$.

In Table 8 some irradiation conditions and maximum target beam currents are reported. The irradiation time is usually limited to one or two half-lives of the ^{201}Pb (9.4 h). The large differences between the maximum allowable

TABLE 8. IRRADIATION CONDITIONS AND MAXIMUM TARGET BEAM CURRENTS

Organization/ country	Type of cyclotron	I_{\max} (μA)	I_{opt} (μA)	I_{coll} (μA)	T (μm)	A ($^{\circ}$)	Remarks
CNEA Argentina	CP42	150	120	15	80	7	No wobbling
NRCAM, Islamic Republic of Iran	Cyclone-30	270	200	80	78	6	Circular wobbling
I.N.P., Kazakhstan	U-150M	150	125	—	160	18	Internal target
K.F.S.H., Saudi Arabia	CS30	125	95	—	40	7	Internal target

I_{\max} = maximum beam current, I_{opt} = beam current used for routine production, I_{coll} = collimator current, T = physical thickness target, A = beam/target angle.

beam currents can be associated with the thickness and geometry of the copper carriers, the thickness of the thallium layer (depending on the beam/target angle), the efficiency of the cooling, and whether or not wobbling is applied.

7.4. TARGET PROCESSING

Irradiated thallium targets can be dissolved in diluted nitric acid, sulphuric acid or concentrated hydrogen peroxide. Using a flow through stripper that allows heating (70–80°C) on the back side, the fast (10–15 min) solubilization in 20–30 mL 0.7N HNO₃ results in co-dissolution of milligram amounts of copper. Consequently, apart from the ²⁰³Tl and the lead and thallium radionuclides formed during the irradiation, the stripping solution will be contaminated with copper and some carrier free ⁶⁵Zn. When time consuming (one hour) dissolution in 4–5M sulphuric acid is done at room temperature, only microgram amounts of copper are dissolved. Though attractive from the analytical point of view, the sulphuric acid dissolution will give rise to a loss of about 10% of the ²⁰¹Pb (and hence of the ²⁰¹Tl) during the solubilization step. Heating to 60–70°C and vigorous stirring will increase the dissolution rate but will also result in greater copper and ⁶⁵Zn contamination. Appropriate chemical processing is then a must. The dissolution in H₂O₂ is

done by drop by drop moistening of the thallium, giving rise to a violent auto-catalytic oxidation reaction resulting in the formation of a dark brown to black slurry of a thallium and hydroxide oxide mixture. The latter are readily dissolved and the thallium is reduced to the monovalent oxidation state by the addition of hot 1N perchloric acid.

Chemical processing involves two steps, the fast separation of ^{201}Pb from irradiated and dissolved ^{203}Tl target material (chemistry-1) at the end of bombardment and the separation of non-carrier added ^{201}Tl from the remaining ^{201}Pb (chemistry-2) after an appropriate grow-in period (25–35 h). It should be noted that only about 90% of the ^{201}Pb has decayed into ^{201}Tl at the moment. Re-extraction of ^{201}Tl from the resulting lead solution after a second decay period may increase the ^{201}Tl yield by more than 5%.

Many radiochemical procedures for the preparation of good quality ^{201}Tl bulks are available from the literature. For Chemistry-1 precipitation (PbSO_4 after addition of lead carrier), co-precipitation (with $\text{Fe}(\text{OH})_3$, SrSO_4), anion and cation exchange chromatography and adsorption chromatography are frequently applied. Chemistry-2 often involves a two step separation: chromatography for the separation of monovalent ^{201}Tl after reduction (SO_2) from the ^{201}Pb bulk (upon decay of ^{201}Pb into ^{201}Tl a mixture of mono and trivalent thallium is obtained) and chromatography and solvent/solvent extraction of trivalent (oxidation with KBrO_3 , O_3 gas) ^{201}Tl . Upon reduction (SO_2 , NaHSO_3), back extraction is performed followed by ^{201}Tl bulk conditioning (pH and isotonicity adjustment, sterility). Some examples are summarized in Table 9 (dissolution, chemistry-1) and Table 10 (chemistry-2 and typical yield).

TABLE 9. DISSOLUTION AND CHEMISTRY-1 PROCEDURES IN ^{201}Tl PRODUCTION

Organization	Dissolution of ^{203}Tl	Chemistry-1
NRCAM	0.7N HNO_3	Precipitation PbSO_4
VUB	Flow through stripper at 60–70°C	Redissolution in EDTA (pH = 5) Cation exchange to remove co-precipitated ^{203}Tl $^{201}\text{Pb}/\text{EDTA}$ bulk EDTA solution decay
CNEA	0.7N HNO_3 Bulk dissolution at room temperature	Cationic exchange 0.1M HNO_3 ^{201}Pb decays on column
I.N.P.	5M H_2SO_4 Bulk dissolution at 80°C	Co-precipitation with SrSO_4 ^{201}Pb decays adsorbed at precipitate

TABLE 10. CHEMISTRY-2 AND TYPICAL ²⁰¹Tl YIELDS

Organization	Chemistry-2	²⁰¹ Tl yield
NRCAM	Reduction SO ₂ , cation exchange (0.1M EDTA)	
VUB	Cation exchange, elution with HCl(6N) ²⁰¹ Tl (6N HCl), Oxidation (O ₃), Extraction DIPE, Reduction (SO ₂), back extraction 0.01M HCl	1–1.5 Ci
C.N.E.A.	Elution of ²⁰¹ Tl (0.1M HCl)	1.2–1.6 Ci
I.N.P.	Redissolution (6M HCl), Oxidation (KBrO ₃) Solvent extraction (Butyl–acetate) Reduction (H ₂ SO ₃), back-extraction (0.01M HCl)	300 mCi

7.5. QUALITY CONTROL

7.5.1. Target quality control

The new electroplating technology from alkaline EDTA solutions allows the production of high quality ²⁰³Tl targets that do not require any post-plating mechanical smoothing. The homogeneity of the thallium layer, determined by micrometry (12 measurements spread over the surface area) is acceptable. An SD on the mean thickness of less than 5% is observed. The SAG (visual microscopic inspection, X250 SEM shots) is excellent as compared with that of targets plated from acid plating baths. The granulometry obtained may be used as a reference for future thallium plating techniques. As the melting point of thallium metal is low (303.5°C), the TST should be performed at 270°C. Heating to this temperature in the air, followed by submersion in cold water (15°C) and multiple bending, does not give rise to crater formation or to peeling off. This proves that neither plating additives (EDTA, BRIJ) nor occlusions are present in the thallium metal lattice, and not at the thallium/copper carrier interface.

7.5.2. Thallium-201 quality control

The ²⁰¹Tl bulk produced by using the new plating technology meets all chemical, radiochemical and radionuclidic quality criteria required by the pharmacopoeia.

7.6. CONCLUSIONS OF SECTION 7

The production of ^{201}Tl by 28.5 MeV proton irradiation of enriched ^{203}Tl (>98%) at higher beam currents (>200 μA) requires high quality electroplated targets.

A new plating/recovery technology allows both the simple electrochemical recovery of the irradiated target material and the simultaneous preparation of as many as eight targets that meet all target quality requirements. The homogeneity of the deposits is better than 5%. An excellent SAG that can be used as an ultimate reference is obtained and the TST is satisfied to 270°C.

The plating is carried out from alkaline (pH > 12.5) 0.5M EDTA solutions containing 0.2% BRIJ-35 surfactant and 1% hydrazine anodic depolarizer. The cylindrical plating geometry involving vigorous bi-directional stirring and the application of an asymmetric chopped bipolar sawtooth as plating waveform, results in a deposit that requires no post-plating mechanical smoothing.

As the plating CE is 100% up to a plating current density of 8.5 mA/cm², time controlled electrolysis is possible when the thallium concentration is such that at the end of a plating batch the thallium concentration is not less than the Cc. Simple replenishment of the partially depleted plating solution with target material allows a plating bath to be reused as many as 10 times.

To take full benefit of the target quality, the irradiation should preferably be performed in a 6° beam/target geometry, in which case the physical thickness of the target layer can be limited to 78 μm . Combined with a proper choice of coolant channel geometry and coolant flow rate, resulting in full turbulent coolant flow, an 11.69 cm² target layer can withstand a 300 μA current burden without loss of target material when appropriate circular wobbling is applied.

Enriched ^{203}Tl can be quantitatively and quickly recovered by CCPE from partially depleted plating baths, or from recovery solutions obtained after acid dissolution (H_2SO_4 , HNO_3) of irradiated targets and separation of ^{201}Pb . In a first step, copper impurities are collected from an acidified solution over 2 h ($\text{H}_2\text{SO}_4/\text{HNO}_3$, pH = 1.5–3) on a copper gauze electrode, the potential of which is set at -0.630 V against an Hg/HgSO₄-sat K₂SO₄ reference electrode. Upon addition of NaOH, EDTA, hydrazine and BRIJ-35 so that the composition of the solution approaches that of the bath used for target plating, ^{203}Tl is recovered on a platinum gauze electrode at -1.62 V against the reference. Finally, upon addition of a large excess of NaOH and gram amounts of zinc carrier, ^{65}Zn is collected on the copper electrode by CCE (30 A) electrolysis. The ^{203}Tl can be readily dissolved in diluted sulphuric acid containing minor amounts of hydrogen peroxide. From this solution, a new plating bath can be

prepared by addition of appropriate amounts of NaOH, EDTA, BRIJ and hydrazine.

The industrial yield of ^{201}Tl was increased by as much as 20% using the new plating technology, and no losses of enriched target material were reported.

8. PLANNING AND OPTIMIZATION OF IRRADIATION CONDITIONS FOR RADIOISOTOPE PRODUCTION USING NUCLEAR DATA

8.1. INTRODUCTION

Nuclear data play an important role in determining the optimal conditions and irradiation parameters for producing the required quality of radioisotope. After preparing a proper target and designing a suitable irradiation set-up, the irradiation process itself should be optimized. This is most important in the case of targets having more than one stable isotope on which nuclear reactions can take place, resulting in radioactive isotopes other than the intended ones. To be able to determine the best irradiation process, the relevant accurate nuclear data for each of the nuclear reactions in question should be available.

Production of radioisotopes in a small cyclotron always requires optimization because of the limited bombardment energy and/or the contaminating side reactions which interfere. The required radionuclide purity level frequently requires highly enriched isotopes, and therefore expensive target materials. Knowing the excitation function of the contributing reactions, an optimization calculation can be performed. Optimization of a production process means choosing a production route that is economical and fulfills the requirements of a high quality product for nuclear medicine. It is always recommended that an a-priori calculation be made to determine the best target and irradiation parameters, bombarding particle and energy, target thickness and irradiation time. For these calculations a database is needed containing the relevant information about the nuclear reactions (main and side reactions) involved in the production process. The required basic information includes the excitation function of the reactions and the stopping parameters of the target material for the available bombarding particles and in the irradiation energy range.

To be able to perform the necessary calculations to optimize the production route, experimental cross-section data were collected from published literature and measured to set up a complete database. Where no experimental data were available the results of model calculation were used instead.

8.2. CALCULATION OF THE TOTAL ABSOLUTE ACTIVITY

A simple calculation tool was developed to calculate the absolute activity and activity ratios of different radioactive products at any target enrichment, and irradiation parameter combinations concerning a single production route.

The required input parameters are:

- (a) Target composition (the absolute ratio of each of the stable isotopes in the target normalized to 1);
- (b) Irradiation parameters:
 - Type of bombarding particle (proton, deuteron),
 - Irradiation time (h),
 - Bombarding beam current (μA units),
 - Primary on target bombarding particle energy (MeV),
 - Outgoing bombarding particle energy (MeV),
 - Cooling time (h).

The use of this calculation tool makes it possible to also estimate the expected activity when all the parameters are fixed and in this way it is suitable to also plan irradiation of a production process.

8.3. PRODUCTION OF IODINE RADIOISOTOPES ON Te OR TeO₂ TARGETS

Production of iodine radioisotopes in a small cyclotron with an acceptable radionuclide impurity level requires highly enriched and expensive Te targets. The activity of the produced iodine radioisotope and the radio-purity level can be calculated from the relevant data of excitation functions, the thickness of the target or the energy range used at irradiation and finally the isotopic composition of the target. Experimental cross-section data of proton and deuteron induced nuclear reactions published earlier was collected and/or measured on all the stable isotopes of tellurium. In addition, theoretical calculations were made using the Alice-IPPE computer code. In the case of ¹²³I

and ^{124}I products, the collected experimental data were critically evaluated and recommended cross-sections were deduced for the investigated reactions [15]. In those cases where only limited experimental data were available, only a spline fit was applied and a smooth curve was calculated over the experimental data. For reactions on which no experimental data were published, the results of the Alice-IPPE theoretical model calculation were used. Based on the available cross-section data, yield was calculated and tabulated for all the contributing reactions and it was applied to estimate the activity produced during the production process.

By using this calculation tool it is easy to estimate the amount of main iodine isotope which will be produced under realistic production circumstances as well as the amount of other iodine radionuclides produced in any side reaction. The amounts of radionuclides produced are given in MBq and also in μCi activity units with correction for decay during irradiation and decay after irradiation during any cooling or chemical processing time. It should be noted that in those cases where no experimental data were available the result of Alice-IPPE calculation was used. Generally, the theoretical calculations result in an excitation function having a higher maximal value than the experimental one. Consequently, the estimated contribution of a reaction calculated using the theoretical cross-sections is an upper limit. The most probable contributing reactions are (p,n), (p,2n) and (p,3n) for proton irradiation, and (d,n), (d,2n), (d,3n) and (d,4n) for deuteron irradiation.

Assuming, as an example, an enriched ^{123}Te target (^{120}Te <0.1%, ^{122}Te 1.6%, ^{123}Te 95.6%, ^{124}Te 2.7%, ^{125}Te <0.1%, ^{126}Te <0.1%, ^{128}Te <0.1% and ^{130}Te <0.1%), 15 MeV bombarding proton energy, a target thickness which causes the outgoing energy to be 5 MeV, then 188 MBq (5.1 mCi) of ^{123}I can be produced in the (p,n) reaction during 1 hour of bombardment at 1 μA . The corresponding ^{124}I activity is 0.45% at EOB that increases to 0.71% 10 h after the end of bombardment. The amount of ^{124}I produced depends on the quantity of ^{124}Te and ^{125}Te present in the target material.

On the same enriched ^{123}Te target using 15–5 MeV deuteron bombardment, 257 MBq (6.9 mCi) of ^{123}I can be produced via the (d,2n) reaction during 1 hour of bombardment at 1 μA . The corresponding ^{124}I activity is 2.21% at EOB and increases to 3.47% 10 h after the end of bombardment.

When using a somewhat cheaper enriched ^{122}Te target (^{120}Te 0.5%, ^{122}Te 95.6%, ^{123}Te 3.5%, ^{124}Te 0.44%, ^{125}Te <0.1%, ^{126}Te <0.1%, ^{128}Te <0.1% and ^{130}Te <0.1%), 86 MBq (2.33 mCi) of ^{123}I activity can be reached by means of the (d,n) reaction during 1 hour of bombardment at 1 μA , calculated for EOB in the 15–5 MeV energy interval. The corresponding ^{124}I activity is 0.34% at EOB, increasing to 0.54% 10 h after the end of bombardment. We can conclude that the (d,n) reaction on ^{122}Te can provide lower activity but cleaner ^{123}I than the

(p,n) reaction on ^{123}Te , which is somewhat more expensive than the ^{122}Te target material.

On an enriched ^{124}Te target ($^{120}\text{Te} < 0.1\%$, $^{122}\text{Te} < 0.1\%$, $^{123}\text{Te} 2.0\%$, $^{124}\text{Te} 95.7\%$, $^{125}\text{Te} < 12.0$, $^{126}\text{Te} < 0.1\%$, $^{128}\text{Te} < 0.1\%$ and $^{130}\text{Te} < 0.1\%$), 715 MBq (193 mCi) of ^{123}I activity can be achieved in the (p,2n) reaction during 1 hour of 1 μA bombardment calculated for EOB in the 30–19 MeV energy interval. The corresponding ^{124}I activity is 1.64% at EOB, increasing to 2.58% 10 hours after the end of bombardment.

8.4. PRODUCTION OF THE ^{103}Pd RADIOISOTOPE

The ^{103}Pd radioisotope can be produced by accelerators with proton and deuteron bombardment of an Rh target. Rhodium has one stable isotope on which the main reaction and some side reactions can take place during bombardment. Similarly to iodine production, a calculation tool was developed to calculate the ^{103}Pd activity and the activity of the other simultaneously produced Pd and Rh radioisotopes. The cross-sections of the relevant reactions were used to estimate the level of the contaminating radioisotopes. The cross-sections were determined experimentally using the stacked target activation technique. Results for proton and deuteron bombardment were published separately [16, 17]. Statistical spline fit was applied over the experimental data and the result was used for calculating tabulated yield for each of the reactions.

Using proton bombardment on a rhodium target with a thickness of 20–4 MeV, 11 MBq (298 μCi) of ^{103}Pd can be produced by means of the (p,n) reaction, during 1 h of 1 μA proton bombardment. The product is free of the possibly contaminating ^{101}Pd radioisotope. Applying deuteron bombardment on a rhodium target with a thickness that degrades the 20 MeV bombarding energy to 5 MeV, 21 MBq (574 μCi) of ^{103}Pd can be produced by means of the (d,2n) reaction during 1 h of 1 μA proton bombardment. The product is free of the possibly contaminating ^{101}Pd radioisotope. Above 20 MeV of proton and deuteron bombarding energy the production of ^{101}Pd is energetically possible and therefore with increasing energy the amount of contaminating ^{101}Pd radioisotope increases proportionately.

8.5. PRODUCTION OF THE ^{201}Tl RADIOISOTOPE

High specific activity, carrier free ^{201}Tl radioisotope is produced by proton bombardment of a thallium target followed by chemical separation of radioactive lead isotopes. In order to increase the radionuclide purity of the

^{201}Tl produced, it is necessary to optimize the irradiation parameters. The (p,xn) reactions result in different radioisotopes of lead. The yields of the lead isotopes produced depend on the primary bombarding proton energy and the target thickness. The cross-section of the relevant reaction is used to estimate the level of contaminating radioisotopes. The measured experimental cross-sections were collected and when enough experimental data points were available the data series were critically evaluated and recommended cross-sections were deduced [15].

Using a 99.9% enriched ^{203}Tl target with a thickness corresponding to 28–20 MeV bombarding proton energy, the amount of ^{203}Pb produced in the (p,x) process during 1 h of 1 μA bombardment at EOB is 836 MBq (22.6 mCi). At the same time, $^{204\text{m}}\text{Pb}$ 0.66%, ^{203}Pb 0.66%, $^{202\text{m}}\text{Pb}$ 4.86% and ^{200}Pb 0.04% are produced.

9. POSSIBLE AREAS OF FURTHER RESEARCH

The participants of the CRP discussed possible subjects of future research arising from the results of this project, as well as from the prospective future needs of radionuclides in modern nuclear medicine. These areas were categorized as:

- (a) Gas or liquid targets: This is related to fabricating targets which will withstand higher beam currents ($>100 \mu\text{A}$) for the production of several radioisotopes with an emphasis on the PET isotopes, in particular ^{18}F . One example would be a liquid water target using ^{18}O enriched water with improved cooling and perhaps a new medium. Another would be an ^{18}O enriched oxygen gas target, which can be run at high beam currents and the ^{18}F extracted efficiently from the walls of the target. The technology for irradiation of gas targets at high beam currents could be applied to additional gas targets for the production of other radioisotopes.
- (b) Solid targets for new radioisotopes: The technology that has been developed as part of this CRP can be applied to a wide variety of other metals which in turn expands the number of useful radioisotopes that can be produced at high beam current with solid targets. The products of these targets could be metals, liquids or gases, as has been shown in the current research. The radioisotopes, which might fall into this category, are ^{62}Zn , ^{62}Cu , ^{64}Cu , ^{67}Cu , ^{68}Ge , ^{76}Br , ^{77}Br and ^{124}I . High beam current

targets would allow efficient production of these isotopes in reasonable irradiation times.

- (c) Extension of current research: Current research has shown that the target cooling parameters and beam optics play a very important role in determining the maximum current which may be applied to a target. Significant research could be done on different methods of cooling, different methods of construction and measurement of beam optics at the high beam currents possible with these targets. Irradiation of other physical forms of target materials such as foils and wires needs to be explored as they may simplify the processing of the target if extraction of the product can be made efficient.

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