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Summary

Specifications for isotopic targets of both stable and actinide nuclides for nuclear measurements are very varied and individually very stringent. The majority of the targets are required to be of specified isotopic and chemical purity, well characterised and uniformly deposited over a precisely defined area. Mass assay of actinide targets may be requested to an accuracy of $\pm 0.1\%$, resolutions of <10 KeV FWHM may be requested for infinitely thin alpha sources and the Schmidt and Pleasonton criteria must be met for ^{252}Cf fission fragment energy calibration sources. In order to cater for this wide range of requirements it is necessary to have available many preparation and calibration techniques. Actinide sources with activities in the range $0.01 \mu\text{Ci}$ to several hundred mCi and targets up to a few mg/cm^2 are prepared by one of the following techniques, vacuum evaporation, painting, electrodeposition or the use of TEG as a spreading agent. These targets may be assayed quantitatively by absolute alpha counting and weighing and qualitatively by alpha spectrometry. Stable isotope targets with thickness varying from a few $\mu\text{g}/\text{cm}^2$ to several hundred mg/cm^2 are prepared primarily by vacuum evaporation and rolling. Mass assay is to $\pm 10\%$ or better, the achievable accuracy being a function of substrate and target thickness. Activities of particular interest include the preparation of fission foils to be used in a series of flux inter-comparison measurements being organised by the Bureau International des Poids et Mesures, the preparation of stable isotope alloy foils for Mössbauer studies and the preparation of actinide targets on thin substrates.

Introduction

Well characterised, highly enriched isotopic targets of both stable and active nuclides are a prerequisite for many nuclear data measurements. Correctly designed targets are fundamental to the success of these measurements so it is important that target specifications are considered at the design stage. All too frequently apparatus is built before the target is requested and the resulting compromise may not be entirely satisfactory.

The target facilities at Harwell have been developed in such a way that it is possible to prepare a wide range of 'once off' type targets without any risk of cross contamination. Emphasis is placed on preparing high quality targets to meet customer specifications in all respects, whenever this is feasible. Self-supporting targets of stable isotopes, carbon, uranium and thorium are prepared as well as deposits of both stable and active nuclides on substrates.

In the case of both stable and actinide isotopes, the most highly enriched and therefore the most valuable nuclides for nuclear data measurements are often available in mg or even μg quantities only. Therefore whenever possible the most efficient deposition technique is used. This can lead to a conflict between achieving target specifications and maximum deposition yield, since the best target preparation techniques tend to be the least efficient. Cross contamination of valuable isotopes and samples must at all cost be avoided. Decontamination of equipment used for stable isotope target preparation is time consuming, but by maintaining scrupulous standards of workmanship this can be

achieved comparatively easily. However the problems associated with actinide target preparation are of a different order of magnitude. There are constraints arising from radiological problems associated with handling actinide nuclides. Also more stringent precautions are necessary to avoid cross contamination, particularly when targets in the 100 mCi range are being prepared. Conventional equipment may be adapted for the preparation of stable isotope targets but specially designed equipment is essential for the preparation of actinide targets.

Stable Isotope Targets

The complete range of stable isotope targets with thicknesses varying from a few $\mu\text{g}/\text{cm}^2$ to several g/cm^2 can be prepared, with certain limitations, by rolling and vacuum evaporation techniques supplemented by die compaction, electroplating and sedimentation.

Rolling is used whenever possible for the preparation of self-supporting foils because it minimises waste of valuable isotopes, and when properly executed introduces virtually no contamination. High purity starting material is essential for rolling thin, light tight foils. Impurities, when chemically combined, can cause hardening and embrittlement and as particles can cause pinholing and cracking. Where necessary, reprocessing of the starting material is carried out using methods which involve minimum volumes of reagents with carefully chosen reaction vessels and conditions, for example purification by electrowinning and vacuum distillation. Self-supporting stable isotope foils of thickness from $500 \mu\text{g}$ to several hundred mg/cm^2 prepared by rolling include Cd, Cu, In, Fe, Mg, Mo, Ni, Ag, Sn, Zn. In specific instances it may be possible to prepare thinner foils but these cannot be guaranteed light tight. Recently there has been particular interest in the production of mixed natural and isotopic alloy foils. These alloys are normally produced in 1.5 g batches from weighed quantities of spec pure natural and isotopic elemental powders. The powders are mixed and ground together in an agate pestle and mortar, after which they are transferred to a compaction die which is pressed at $45 \text{ tons}/\text{cm}^2$. The resulting pellets are transferred to an argon arc button producer, which can fuse quantities as small as 50 mg. A water cooled hearth eliminates crucible contamination and also loss of valuable material, which alloys to walls when crucibles are used. The high temperatures used, in excess of 4000°C , improve product homogeneity. A water cooled hammer to flatten the metal bead eliminates quenching operations and helps to improve bead and resultant foil homogeneity. Oxidation is eliminated by fusing in an argon atmosphere. During rolling, when annealing is necessary, it is done in an argon-5% hydrogen atmosphere at 800°C . When the foils have been rolled to the correct thickness they are finally annealed. Pd ^{57}Fe alloys are prepared for calibrating Mössbauer equipment.

Iron based alloys containing 0.2 atom per cent of ^{119}Sn with 2 to 6 atom per cent of one of the following elements, Al, Cr, Cu, Ge, V, Au, Ir, Os, Pt, Rh, Si, Ti, W, Zn, As, Pd, Mo, Re, Ni, Co and Ru have been prepared for the investigation of temper embrittlement by Mössbauer Spectroscopy.

Nickel based alloys with 0.2 atom per cent ^{57}Fe and 2 to 5 atom per cent of Cr, V, and Ti, and an alloy of Zr with ^{119}Sn have been prepared for investigations of void swelling behaviour.

Vacuum evaporation¹ is used to prepare stable isotope targets of less than 1 mg/cm^2 thickness. Targets are prepared on substrates or as self-supporting foils. In the latter case, the stable isotope is evaporated on to a backing foil which is subsequently dissolved away, or on to a glass microscope slide coated with release agent from which the stable isotope target is subsequently floated off. This technique produces high quality targets but the efficiency is low and can be $<1\%$ for targets deposited uniformly over a relatively large area. Resistance heated filaments are used for the evaporation because of the small quantities of isotope available. To aid isotope recovery, a glass pipe coated with Zapon is interspersed between the evaporation filament and the substrate. On completion of evaporation the stable isotope deposited on the inside of the pipe is removed with acetone and subsequently recovered.

^{119}Sn targets $400\text{ \AA}$ thick have been deposited on natural Ti and Fe substrates. The ^{119}Sn is then further implanted into the substrate by nitrogen ion bombardment; such targets are used to investigate wear in artificial hip joints. Targets of ^{206}Pb and ^{209}Bi $500\text{ }\mu\text{g/cm}^2$ thick have been strengthened by supporting them with a layer of Zapon. The isotope is evaporated on to a microscope slide coated with CsI, Zapon in methyl ethyl ketone is then sprayed on and allowed to dry. Finally the target is floated off the slide.

Electroplating is little used for preparing stable isotope targets because of the risk of introducing impurities into the target material. However, it is used for coating irregular shaped metal backings. Also single crystals of elemental iron have been coated with ^{57}Fe for Mössbauer studies.

Die compaction is used for the preparation of very thick targets of, for example, boron for use as beam stoppers. Isotopic lithium pellets, including those from synthetic mixtures of ^6Li and ^7Li , contained in holders of the same material have also been prepared from both lithium hydroxide monohydrate and lithium carbonate using this technique.

Targets of refractory and thermally unstable compounds, e.g., rare earth, Ti, Ge and Si oxides are prepared by sedimentation. The oxide is slurried in acetone and added dropwise to a cell, to the bottom of which the substrate is clamped. The acetone evaporates leaving the substrate coated with an oxide deposit.

Actinide Isotope Targets

Well characterised actinide nuclide sources and targets are required for calibrating equipment, for the measurement of nuclear constants, in transmutation experiments as targets for ion bombardment in accelerators, and as neutron dosimeters. The source requirements for these applications are very varied and individually very stringent. The substrate is usually required to be of a particular size and shape. It may be small and fragile, e.g., $6\text{ }\mu\text{in}$ nickel, or alternatively it may be large, e.g., 100 mm diameter, 0.125 mm thick platinum foil. The activity is normally required to be deposited over a precisely defined area which may be a central spot $1\text{--}2\text{ mm}$ diameter, a rectangle, a narrow slit, or very many cm^2 . Criteria for uniformity which must be met are frequently very stringent. The sources must be free from radioactive and non-active contaminants, the isotopic composition must be accurately

specified, and in the case of short half-life nuclides, daughter products must be accurately assayed. Other specifications requested include, mass assay to $\pm 0.01\%$, resolutions of 10 KeV Full Width Half Maximum Peak Height (FWHM), and for Cf^{252} fission fragment energy calibration sources the Schmidt and Pleasanton criteria should be met. Actinide sources can be prepared with activities from $0.0001\text{ }\mu\text{Ci}$ to several hundred mCi and targets for nuclear experiments with deposits up to a few mg/cm^2 .

To cater for such a wide variety of requirements poses many problems. These include handling problems associated with using high specific activity nuclides, the avoidance of cross contamination of valuable isotopes, and not least meeting often near impossible target specifications. Other most important criteria which must be met are that any uncovered source must be safe when handled with appropriate precautions and that thick deposits in the mg/cm^2 range must be smooth and adherent; powdery looking deposits are unacceptable on safety grounds.

Chemical separations have in many cases to be carried out immediately prior to target preparation, either to remove daughter products or non-active impurities present in the solution arising from radiation damage to the containment vessel. To prepare infinitely thin, high resolution actinide sources the starting solution must be chemically clean. Target materials such as ^{242}Cm , ^{235}Np and ^{236}Pu are prepared by reactor or cyclotron irradiation and chemically purified.

The two techniques most regularly used for actinide target preparation at A.E.R.E., Harwell are vacuum evaporation² for the preparation of infinitely thin, 'weightless' deposits, and painting, which in our experience has no equal for the preparation of thick (mg/cm^2) adherent deposits built up by the application of many layers. Electrodeposition is also used specifically for the preparation of sources with active deposits less than 1 cm diameter and activities in the mCi range.

Vacuum evaporated deposits with resolutions of less than 10 keV (FWHM) can be produced without difficulty, as well as deposits of up to $250\text{ }\mu\text{g}$ per cm^2 on either rigid or thin substrates. The essence of the vacuum evaporation equipment is simplicity. The evaporation chamber is readily demountable from a purpose built vacuum rig and a separate chamber is dedicated to each actinide nuclide handled, thus eliminating cross contamination. Vacuum evaporated targets have been prepared from ^{226}Ra , ^{229}Th , ^{230}Th , ^{232}Th , ^{231}Pa , ^{232}U , ^{233}U , ^{234}U , ^{235}U , ^{236}U , ^{237}Np , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu , ^{241}Am , ^{243}Am , ^{242}Cm , ^{244}Cm , and ^{252}Cf to a wide range of specifications.

The majority of actinide sources are prepared by vacuum evaporation and of these, very many are on thin substrates. For many experiments a thin light element substrate is desirable. However this must be related to other factors of equal importance such as active deposit thickness and area, unsupported substrate area and specific activity of the requested nuclide. Additional factors which have to be considered are the safety of the completed target and its anticipated shelf-life. Inevitably the final target is a compromise between that which is desirable and that which it is feasible to prepare.

The feasibility or otherwise of preparing actinide deposits on thin substrates is very finely balanced between the thickness and material of the substrate, the unsupported area, the active area, and the specific

activity of the nuclide in question. To a rough approximation it is desirable that the deposit thickness does not greatly exceed the substrate thickness. 6 μ m nickel is the most extensively used thin substrate for actinide targets, since this is the thinnest material which can be guaranteed light tight. However, deposits can be prepared on 50 μ g/cm² thick VYNS and carbon substrates, but these are very fragile and do not travel well. 1.5 mg/cm² Melinex is a satisfactory thin substrate and has a comparatively long shelf-life. A ²²⁶Ra target 30 μ g/cm² was prepared comparatively easily over an active area 1 x 3 mm on 50 μ g/cm² carbon backing and had a shelf-life of many years. However all attempts to prepare a similar target of 5 mm active diameter failed. This highlights the importance of considering target feasibility when designing an experiment. Similarly, whereas it is possible to deposit 200 μ g/cm² ²³⁵U over 10 mm active diameter on 6 μ m nickel backing foil having an unsupported diameter of 18 mm, at 15 mm active diameter it is difficult to achieve a deposit thickness of 150 μ g/cm². For high specific activity nuclides, safety and radiation damage to the substrate tend to be the limiting factors. For example, ²³⁹Pu is usually limited to 50 μ g/cm² on safety grounds. 10 μ Ci ²⁵²Cf sources have been prepared on 6 μ m nickel and experience has shown that at this activity level an active area 4 mm diameter will have a longer shelf-life than a 10 mm diameter active area. An extensive range of sources has been prepared on 6 μ m nickel including ²³²U, ²³⁴U, ²³⁵U, ²³⁸U, ²³⁸Pu, ²⁴⁰Pu, ²⁴¹Am and ²⁴³Am at activity levels of 10 μ Ci, active deposit diameter 10 mm, and unsupported aperture 20 mm diameter.

Vacuum evaporation has been used to prepare ²⁴²Cm sources on rigid substrates at activity levels of several hundred mCi using equipment installed in a glove box.

Painting³ continues to be the method used exclusively for the preparation of uniform deposits from 250 μ g/cm² to 8 mg/cm². The active area is defined by a mask. ²³⁸Pu and ²³¹Pa targets with activity levels up to 350 mCi deposited over many cm² on platinum and stainless steel backings have been prepared by painting. These sources were required for irradiating biological samples where uniformity and safety were the most important criteria.

Two sets of fission foils, one of >99 per cent enriched ²³⁵U and one of highly depleted ²³⁸U are currently being prepared for two standard fission chambers, to be used in a series of flux intercomparison measurements being organised by the Bureau International des Poids et Mesures in Paris (B.I.P.M.). The backing foils are 89 mm diameter, 0.127 mm thick platinum, with activity deposited over 76 mm diameter on each side of the foil. Five foils of each nuclide are being coated to a thickness of 100 μ g/cm² and five to a thickness of 500 μ g/cm². When preparing double-sided foils a Teflon liner is fitted to the painting mask to avoid abrasion of the underside of the foil. The uniformity of each deposit is required to be such that the average deposit over any area of one square centimeter does not differ from the average for the whole deposit on that particular side by more than $\pm 4\%$. Each foil is being autoradiographed and comparisons of deposit uniformity are being made with a densitometer. Uniformities of $\pm 2\%$ can be achieved on carefully prepared painted deposits. Quantitative checks on uniformity will also be made by alpha counting. The mass of each deposit is required to be known to better than $\pm 0.5\%$. The deposits are being assayed by direct weighing and by alpha counting, using a purpose built medium geometry counter; indications are that the agreement between weighing and counting is within the

required accuracy. Finally, one foil from each set will be assayed destructively and the uranium determined by controlled potential coulometry and isotope dilution mass spectrometry. The isotopic composition of the foils is being determined mass spectrometrically. ²³⁷Np deposits for use in personnel fast neutron dosimeters have been prepared on aluminium substrates by a combination of anodising and painting. The ²³⁷Np was required to be deposited uniformly over the entire surface of one side of 0.010 inch thick aluminium foil 30 x 30 mm. A large number of these deposits were needed, and the criteria for uniformity and total activity were that these should not exceed $\pm 5\%$ over the entire batch of deposits. High purity ²³⁷Np containing <15 p.p.m. of nuclides with large thermal cross sections was essential. Additionally the deposits had to be sufficiently adherent so that there was no risk of contaminating the wearer or the track recording film. The aluminium backing foil was surface anodised then fired at 400°C to form a hard but porous aluminium oxide layer of the order of 1 μ m in thickness. This absorbed the paint solution very uniformly. The deposits were prepared in batches of six by painting an area 90 x 110 mm and punching out 30 x 30 mm squares. The foils were very abrasion resistant and therefore inherently safe.

²⁴⁴Cm and ²⁴¹Am targets with activities in the range 1-5 mCi deposited over 1 cm active diameter have been prepared by cathodic electrodeposition on to platinum substrates from ammonium formate buffer solution^{4,5}. Deposition yields were of the order of 50% with a total nuclide recovery of approximately 90%.

To prepare sources where high activity levels cause serious handling problems or where the quantity of activity available is so small that losses cannot be tolerated, tetra ethylene glycol (TEG) is used as a spreading agent⁶. The activity is deposited directly on to the substrate and a drop of TEG is added; when the mixture is warmed and dried the TEG polymerises and a uniform film is produced. The TEG is finally removed by ignition to 800°C leaving a uniform adherent deposit.

In the mCi range 'sealed' sources are sometimes requested. However, this is a misnomer in so far as experimental targets are concerned, since typically, absolute minimum energy loss in the window material is required. Energy degradation in a variety of window materials and their suitability for use under vacuum have been examined. Nickel foils are particularly uniform and the emergent alpha spectra are good. However, nickel is not radiation resistant and deteriorates with time. Our experience suggests that a 'sealed' source with a readily interchangeable window is preferred or, alternatively, an experiment using an actinide source should be designed in such a way that the source can be stored in a 'sealed' safe position and exposed experimentally without a window. Either of these two alternatives avoids the hazard resulting from the window collapsing on to the source.

Calibration

The absolute disintegration rates of single nuclide sources are measured in one of two similar low geometry proportional counters⁷ used for ²³¹Pa⁸ and ²³⁹Pu⁹ half-life measurements. Activities in the range of 0.1 μ Ci to 0.1 mCi can readily be measured to an accuracy of $\pm 0.1\%$, and to $\pm 1\%$ for sources > 0.01 μ Ci and < 0.1 μ Ci. Sources at the level of mg/cm² are estimated by weighing, and whenever possible, checked by low geometry alpha counting.

To determine accurately the individual constituents of an alpha source consisting of several nuclides, the total disintegration rate is measured in a low geometry counter and a qualitative analysis made using a premium series surface barrier detector, or for very low activity sources a gridded ion chamber may be used. Alternatively, the absolute disintegration rate of an individual nuclide in a spectrum from a thin source with associated daughter products (e.g. ^{228}Th) can be measured directly by feeding the output from a low geometry counter with a surface barrier detector into a multichannel pulse height analyser. The accuracy of these measurements is better than $\pm 1\%$ for activities in the range $0.01 \mu\text{Ci}$ to 0.1 mCi .

Fig. 1 shows a typical alpha spectrum from a ^{238}Pu source. ^{252}Cf fission fragment energy spectra are measured using a surface barrier detector and a typical spectrum conforming to the Schmidt and Pleasonton Criteria is shown in Fig.2.

Fig. 1. Alpha energy spectrum from a vacuum sublimed Pu-238 source on Mylar backing.

Fig. 2. Fission fragment energy spectrum from a vacuum sublimed Cf-252 source.

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