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Source Preparations for Alpha and Beta Measurements

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Abstract

Regarding alpha particle emitters subject for environmental studies, electrodeposition or co-precipitation as fluorides are the most common methods. For electro deposition stainless steel is generally used as cathode material but also other metals such as Ni, Ag, and Cu showed promising results. The use of other anode material than platinum, such as graphite should be investigated. For other purposes such as optimal resolution other more sophisticated methods are used but often resulting in poorer recovery.
For beta particle emitters the type of detection system will decide the source preparation. Similar methods as for alpha particle emitters, electrodeposition or precipitation techniques can be used. Due to the continuous energy distribution of the beta pulse height distribution a high resolution is not required. Thicker sources from the precipitates or a stable isotopic carrier can be accepted but correction for absorption in the source must be done.

Keywords

Source preparation; Energy resolution; Alpha spectrometry; Beta measurement; Electroplating; Precipitation; Anode; Cathode

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Report from the NKS/BOK-1.1 project,
Laboratory Measurements and Quality Assurance

Contents

Summary

Under the Nordic Nuclear Safety Research programme, subproject BOK-1.1: Laboratory measurements and quality assurance, different methods for source preparation for alpha and beta measurements were investigated. This consisted mainly of a literature research but also experimental work was carried out.

Recent development of detector systems and environmental assessment of both alpha and beta particle emitters in the environment has created a need for rapid and reliable methods for source preparation.

For preparing alpha sources the evaporation technique was often used in the beginning of for example analyses of alpha particle emitters in environmental samples. The possibility to perform sources of actinides by electrodeposition is known for over 40 years and has been applied to environmental assessment and this technique gives a better energy resolution. Precipitation as fluorides with rare earths provides generally a sufficient resolution and has the advantage that it is rapid and the yield is high. Molecular plating can be applied also to elements such as radium, which is not possible by normal electrodeposition. Other techniques such as vacuum sublimation and electro spraying are used for optimal energy resolution but the recovery is small.

For beta particle emitters the most common source is prepared for liquid scintillation. For other counting systems such as GM-counters similar techniques as for alpha particle emitters, electro deposition and precipitation techniques are used.

Introduction

The problems associated with alpha and beta spectrometry/measurements can be divided into three main categories,

a) those associated with the detector and electronic equipment,
b) the radiochemical separation technique,
c) those associated with the source preparation.

The relative importance of these factors depends on the type of measurement undertaken. However in many cases the source thickness is the limiting factor, whatever type of detector system is selected. Sophisticated software for resolving poor spectra can not replace good sources. Poorly resolved spectra will also seriously affect the analytical quality control. The source should have a minimum of foreign material present and should be capable of withstanding careful handling. Normally an infinitely thin source on a flat substrate is desired. Other configurations have generally not been considered.

Even if the thickness of the source is homogenous over the active surface, the energy resolution, Full With at Half Maximum, will increase due to energy straggling. Alpha particles will traverse different thickness depending on the pathway and the distance to travel for the particles will differ with the angle to the detector.

A review of source preparation for alpha spectrometry was done in 1984, (Lally and Glover). Most of original work was developed in the 1970ies-1980ies and work during later years is a further development of what was achieved then. An example of degradation of alpha spectra with increasing source thickness is shown in Fig. 1.

Yakolev G.N., Chulkov P.M., Dedov V.B., Kosyanov V.N., Sobolev Y.P., 1957.
The preparation of thin films of plutonium, americium, and curium by an electrolytic method.
Atomnaya Energiya, 1, p. 131.

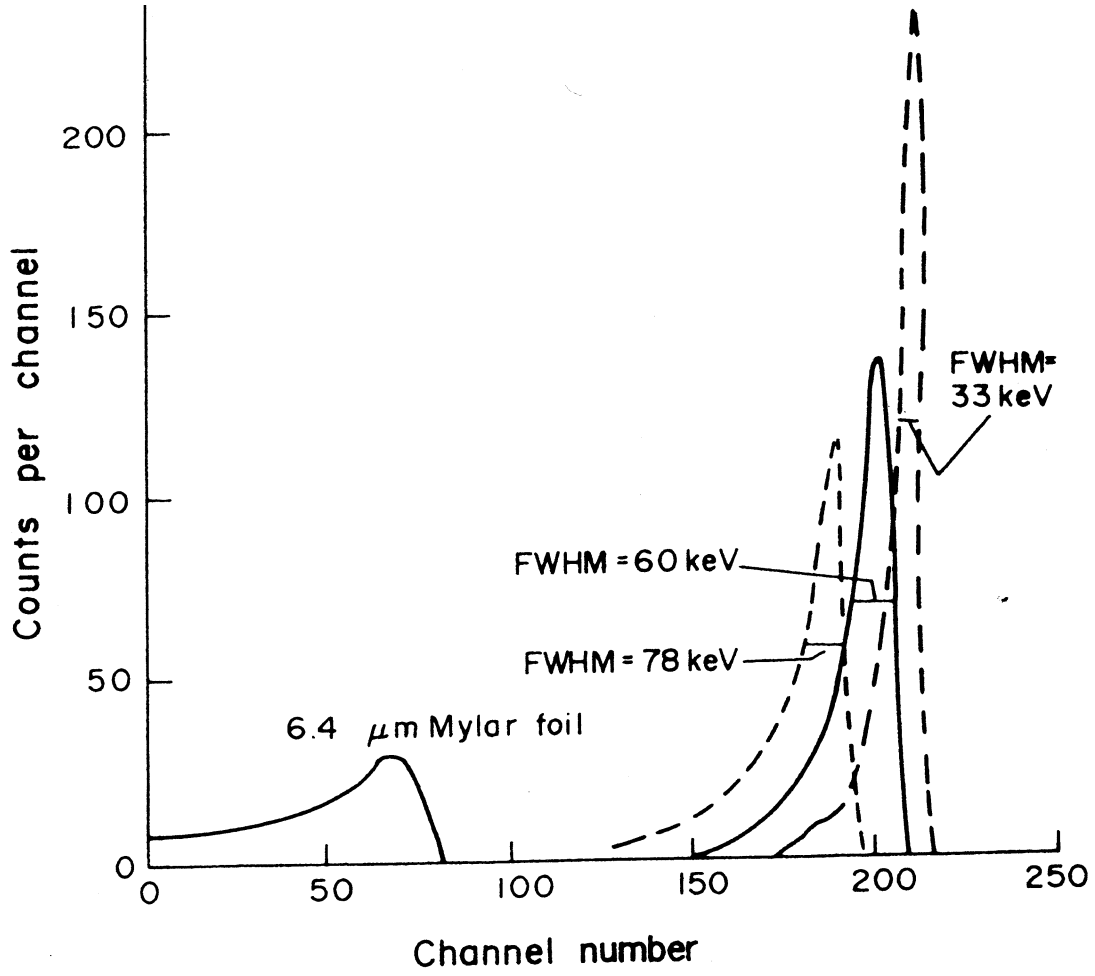

Fig. 1. Degradation of α-particle spectrum from a $^{239+240}$Pu source with varying absorber thickness (From Holm, 1984).

Different Methods for Source Preparation for Alpha Spectrometry

Evaporation

The evaporation technique was often used in the beginning of for example analyses of alpha particle emitters in environmental samples. This technique is rapid and is quantitative. An aliquot is evaporated onto a solid substrate and evaporation is performed using a heat source or an IR lamp.

Direct evaporation tends to give non-uniform sources. Spreading agents such as TEG, tetraethylene glycol, can be used to overcome this phenomenon.

The quality of the source will depend on the solid content of the solution. Spreading over a large surface will improve resolution. Sources of good quality can be obtained by using glazed porcelain or quartz as support since heating the tiles at temperatures up to 850 ^{o}C can eliminate many undesired elements. (Miguel *et al*, 1984).

Electrodeposition

This is the most common method today for determining α-particle emitters in environmental samples after radiochemical separation. The method can be divided into electroplating from aqueous solutions and molecular plating. Such methods for producing thin films of actinides were published already in the 1950ies (Yakolev, 1956, Ko, 1957).

Electroplating

The mechanism for electroplating from aqueous solutions is still not very well understood. It is believed that either the hydroxides together with minor quantities of the anode material is plated out as hydroxides or a reduction to the metal takes place with subsequent deposition on the cathode. Mot likely the actinides are co precipitated with platinum from the cathode as hydroxides (Ferrero Calabuig *et al*., 1998, Weber *et al*., 1999).

As cathode polished stainless steel is generally used today. Platinum as cathode has no major advantage except if the activity is washed off for reprocessing or for ex. ^{241}Pu determination after build up of ^{241}Am. Stainless steel will give iron and nickel in the solution, which again have to be separated before replating.

The most common electrolytes are ammonium salts, sulphate, chloride, oxalate or formate (Talvitie, 1972, Lee et. al., 1994, Lee and Pimpl, 1999). The plating normally takes 1-4 hours at a current of 300 mA cm^{-2}. Several samples can be electroplated at the same time and cells are normally in serial connection. The current is set constant and a tension of about 5-10 V per cell will be required. An addition of some sodium sulphate prior to the final electrolyse preparation diminish losses (Hallstadius, 1984).

The pH should be carefully adjusted to about 2, which seems to be more important for certain radionuclides such as americium and thorium. Rotating anodes and cooling of the electrolytes are sometimes used (Becerril 1993), but there is no evidence of higher recoveries or improved resolution by such actions. Fig 2. shows an example of an electrodeposition cell and Fig. 3 a pulse height distribution of thorium with ^{229}Th as yield determinant, separated from an environmental sample

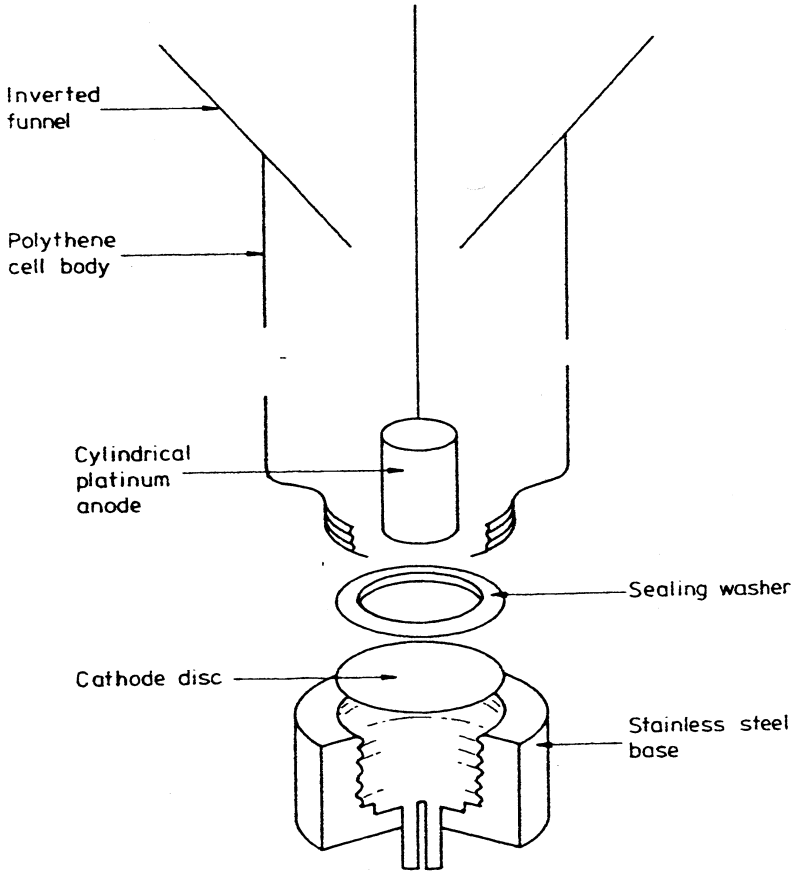

Fig. 2. Example of the construction of an electro-deposition cell (From Lally and Glover, 1984).

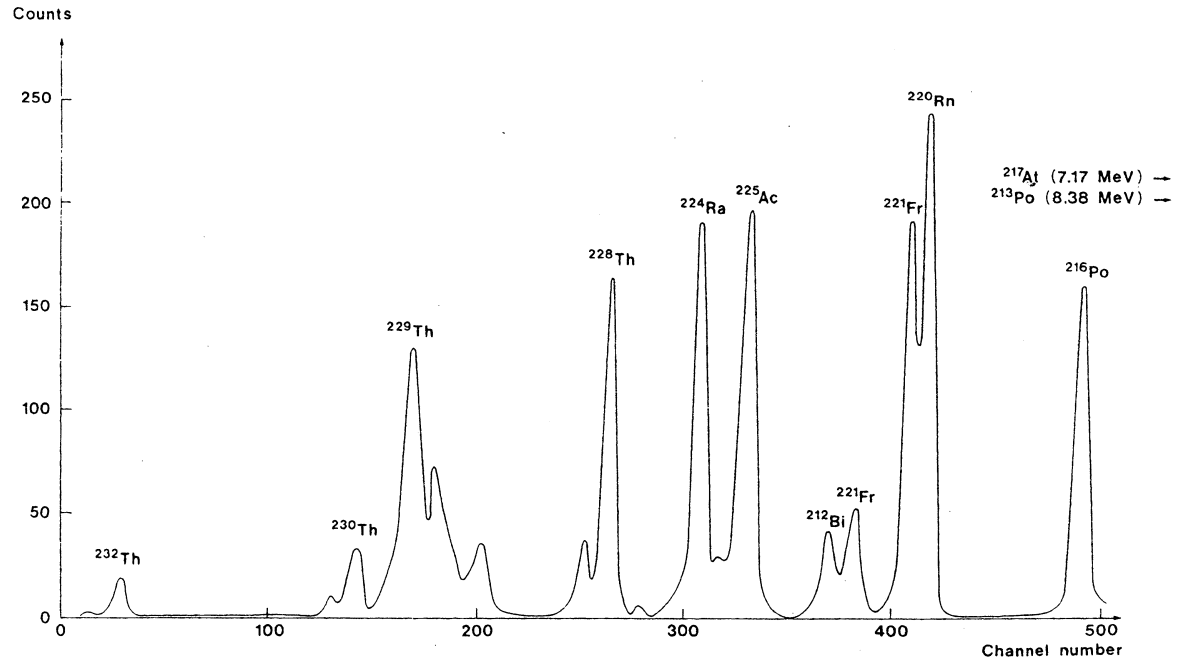

Fig. 3. Alpha spectrum after thorium separation from sea water and electrodeposition using ^{229}Th as yield determinant, with a surface barrier Si-detector (From Holm, 1984).

Molecular Plating

Molecular plating is carried out by passing a high voltage (100 - 500V) low current supply through an organic solution of a molecular species. Electrodeposition from nitric acid-isopropyl alcohol also on aluminium has been performed. The deposit is thought to have the same molecular form as the anion initially present in the solution. Electroplating can also be performed by mineral acid-alcohol (or other polar solvents) solutions. Even in 20 minutes also radium will plate from such a solution (Roman, 1985).

Vacuum sublimation

When a compound is heated to high temperature (2400 ^{o}C) in vacuum, it is vaporised and can be sublimed to a substrate. The evaporation is achieved by passing a current through a filament, tungsten, tantalum or platinum, or by radiofrequency heating. The technique is more appropriate to metrology applications than environmental radionuclide determinations.

Precipitation

Actinides can be precipitated as hydroxides, fluorides or sulphates. A sequential combination of the precipitates can be used for elimination of interfering elements. The most common technique for the final source preparation is to use a co- precipitation of actinides in the +3 or +4 oxidation states with rare earths such as Ce or La. A small amount (10 – 50 μg) of for example La is added to the sample and precipitation is done by hydrogen fluoride (Joshi, 1985, Luskus, 1998). The precipitate is filtered through a filter paper and after drying, the filter is mounted on a counting tray.

Electrospraying.

The sample is dissolved in an organic solvent normally ethyl alcohol. The solution, 100 μl, is transferred to a capillary tube. An electrode is inserted in the solution making the anode. A potential of 8 kV is applied and the organic solution is ejected as a fine spray from the anode to the cathode. Due to evaporation only solid particles reach the anode, which is rotated if a very uniform source is required.

Electrostatic precipitation

This technique is used for environmental air samples of large volumes. The samples can be collected on microsorbant filters and then analysed in a conventional way using radiochemical procedures. Alternatively the sample is collected on large electrostatic precipitator and the sample is counted on a large gridded ionisation chamber. For example several days counting using a tin plate cathode operating at a corona discharge at – 9kV has been reported. This method is mainly used for monitoring/screening purposes.

Spontaneous deposition

The most classical example is the deposition of Polonium on silver or nickel from aqueous solutions (Flynn, 1968). This method is still used for determination of ^{210}Pb and ^{210}Po in environmental samples for dose assessment and dating. Any size of source can in theory be produced. Since ^{210}Po and ^{209}Po are monoenergetic they are used as calibration sources.

Recoil sources

Recoil nuclides can be plated out on surfaces, which can be used as sources especially for retrospective assessment of radon in dwellings (Samuelsson, 1996). The final source will be a

$^{210}Pb/^{210}Po$ source. In this way relatively large sources of glass surfaces such as mirrors, windows, glass frames for photographs etc. can be measured with gridded ionisation chambers.

The phenomenon of recoil daughter products, for example ^{224}Ra from ^{228}Th, on detectors and surfaces during the alpha spectrometric measurement is more a contamination problem than having any practical application.

Different Methods for Source Preparation for Beta Measurements

Electroplating

As a general rule the same methods as for preparation of sources for alpha spectrometry, such as electroplating can be used. This rule is also valid for source preparations for elements decaying by electron capture such as ^{55}Fe and ^{59}Ni, emitting very low-energy photons. Such methods are necessary for good beta spectrometry. Especially if conversion electrons are detected and studied this might be desirable. Examples of this are demonstration of conversion electrons from $^{99}Tc^{m}$ and ^{51}Cr and environmental measurements of ^{63}Ni. For ^{63}Ni there is no suitable radioactive yield determinant and stable nickel is used. This means that the counting efficiency will depend on the yield and how much stable nickel that is used. Fig. 4 shows the counting efficiency of ^{63}Ni as a function of deposition thickness for two types of detectors.

Sources for liquid scintillation

However due to the relatively longer range of beta- compared to alpha particles in material and the fact that the beta spectrum has a continuous energy distribution from zero to a maximal energy, other counting systems than solid state detectors are generally used. We are here mainly thinking of liquid scintillation and GM gas counting. Liquid scintillation will provide some energy resolution even if this is rather poor and identification of end point energies can be done. A couple of beta emitters in the same sample can be measured simultaneously by energy discrimination.

The source is prepared for example dissolving organic extractants such as TBP, TTA etc containing the radionuclide in an organic scintillator. Another possibility is to have the radionuclide in a very small aqueous solution, that is mixed into a gelatinous material together with the scintillating material. In this way very low energy beta particles can be counted since there is hardly any entrance window for creating the scintillation.

Precipitation

For radionuclides emitting beta particles with higher energies precipitation techniques can be used hydroxides, sulphates, oxalates etc. The classical example is separation of ^{90}Y from ^{90}Sr by precipitation with stable yttrium as yield determinant. However the amount of precipitate will always affect the counting efficiency due to the continuous energy distribution of the beta particles.

Sorption techniques

Sorption onto ion-exchangers or activated carbon should be possible, rapid and effective for measurements of example ^{99}Tc but has not yet been employed.

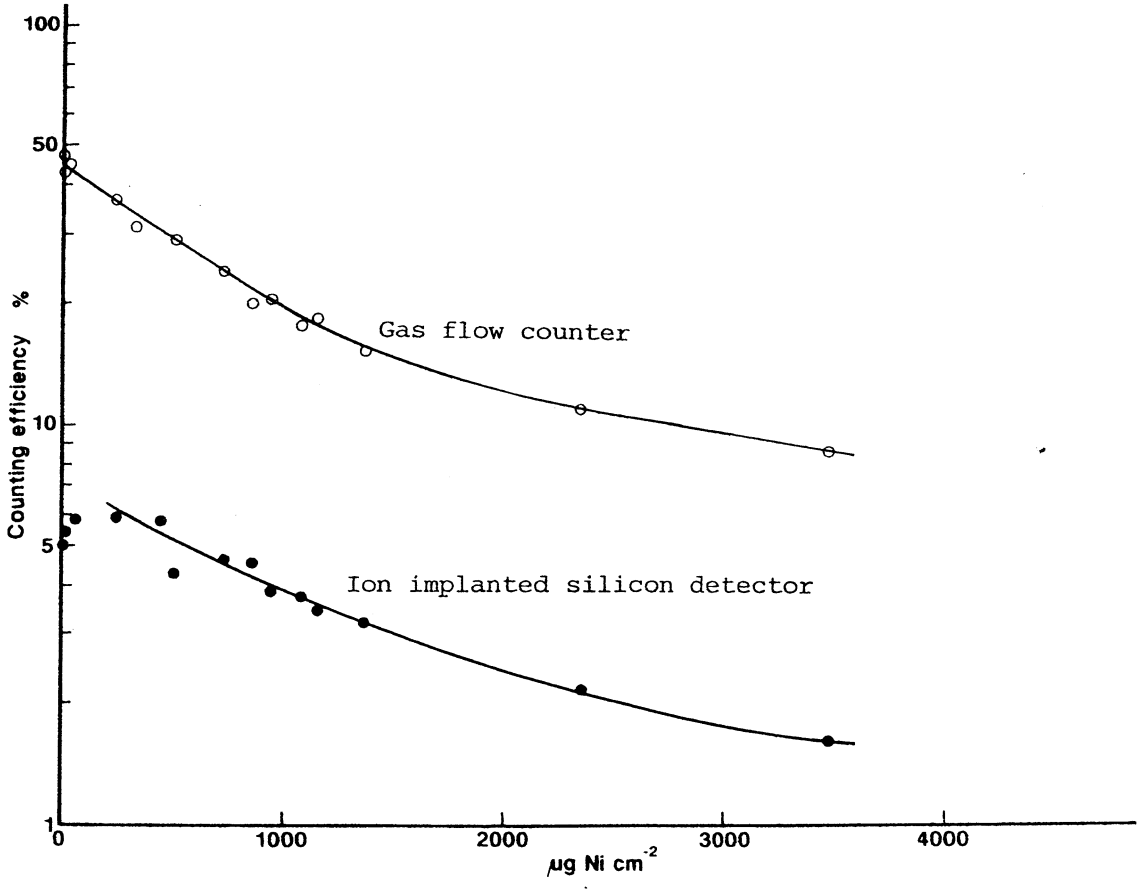

Fig. 4. Efficiency calibration for ^{63}Ni of open gas flow counter and ion implanted silicon detector for different amounts of stable nickel on the source (From Holm et al., 1990).

Identification of Further Work

If we concentrate our efforts to the analysis of alpha particle emitters in environmental samples there are two methods, which are more suitable, and also more used than other methods, electrodeposition and precipitation. For source preparation for calibration purpose when very high yields are not necessary other methods are also applicable. Some of them require sophisticated apparatus and are not convenient for preparation of hundreds or thousands of sources per year.

Precipitation

The precipitation techniques have the advantage over electrodeposition by being more rapid with the expense of some poorer energy resolution. Other precipitation or rather sorption techniques should be tried for example sorption small amounts of activated carbon or ion exchangers.

Electrodeposition

While a large number of different electrolytes have been used relatively few other cathode materials than stainless steel and platinum have been tried for thin source preparations. The possibility for general source preparation from standard solutions using aluminium and copper or polyamide foils coated with conducting layers of gold, platinum or carbon using

molecular plating has been shown (Ingelbrecht et al., 1997). Such methods should be developed further for environmental analysis.

It seems as the only anode material used is platinum and as explained above a small fraction of the platinum is deposited together with the radionuclides. The possibility to use for example graphite or even aluminium should be investigated. The important thing is that the anode material does not deposit on the cathode even if it should be consumed to some extent.

Some preliminary trials were done and the results can be seen from Tables 1a and 1b using the standard method (Hallstadius, 1984). It is obvious that other cathode material such as copper, silver and nickel can be used. The use of graphite as anode seems to be inferior to using a platinum electrode. A general tendency is that thorium is more difficult to electroplate and to some sense also americium. The pH control is more important than for other actinides.

Methods to improve resolution

Other factors affecting resolution than badly conducting sources or source mounts are conversion electrons produced by transitions between α-emitting levels having fine structure. This will also affect evaluation when spectral deconvolution is done.

The possibility to deflect or retard such electrons then not reaching the detector in coincidence with the alpha particle should be investigated.

Table 1a. Electrodeposition of actinides using platinum anode and different cathode material

Platinum anode	**Am, % yield**	**Pu, % yield**	**Th, % yield**	**U, % yield**
Steel cathode	94	100	81	81
Copper cathode	96	100	75	87
Silver cathode	91	100	70	84
Nickel cathode	94	100	52	100

Table 1b. Electrodeposition of actinides using graphite anode and different cathode material.

Graphite anode	**Am % yield**	**Pu % yield**	**Th % yield**	**U % yield**
Steel cathode	27	27	17	39
Copper cathode	10	13	12	96
Silver cathode	35	32	17	98
Nickel cathode	36	35	24	95

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