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Application
of Distillation Techniques
to Radiochemical
Separations

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Application of Distillation Techniques to Radiochemical Separations

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National Bureau of Standards
Washington, D. C.

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Subcommittee on Radiochemistry
National Academy of Sciences — National Research Council

FOREWORD

The Subcommittee on Radiochemistry is one of a number of sub-
committees working under the Committee on Nuclear Science within
the National Academy of Sciences - National Research Council. Its
members represent government, industrial, and university laboratories
in the areas of nuclear and radiochemistry.

The Subcommittee has concerned itself with the preparation of
publications, nuclear education, special problems, and sponsorship
of symposia on selected current topics in nuclear and radiochemistry.
A series of monographs on the radiochemistry of essentially all the
elements and on radiochemical techniques is being published. Initi-
ation and encouragement of publication of articles on nuclear edu-
cation in the areas of chemistry have occurred, and development and
improvement of certain educational activities (e.g., laboratory and
demonstration experiments with radioactivity) have been encouraged
and assisted. Radioactive contamination of reagents and materials
has been investigated and specific recommendations made.

This series of monographs has resulted from the need for compre-
hensive compilations of nuclear and radiochemical information. Each
monograph collects in one volume the pertinent information required
for radiochemical work with an individual element or with a specialized
technique. The U.S. Atomic Energy Commission has sponsored the printing
of the series.

Comments and suggestions for further publications and activities
of value to persons working with radioactivity are welcomed by the
Subcommittee.

N. E. Ballou, Chairman
Subcommittee on Radiochemistry

ABSTRACT

The use of vacuum distillation of inorganic compounds and metals for radiochemical separations is reviewed. A brief description of the experimental apparatus which is most often used for the distillation is outlined. Advantages and disadvantages of the various techniques are discussed. Detailed separation procedures are described for wet chemical distillation separations as well as for separations of daughter radioactivity from target materials by direct heating of the target.

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Application of Distillation Techniques to Radiochemical Separations

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I. INTRODUCTION

An adequate radiochemical separation can be defined as a separation of the radioisotopes of a given element from those of a group of other elements so that maximum purity and yield are obtained in a time which is short compared to the half-life of the desired radioisotope of the element which is separated. The distillation technique has been used less often for radiochemical separations than methods such as extraction or ion exchange; however, in some instances distillation proves to be the best method.

It is the purpose of this report to indicate how well distillation techniques satisfy the requirements of radiochemical separations, and to point out which elements can be conveniently separated by distillation.

In recent years some consideration has been given to the preparation of carrier-free sources by the distillation technique. Therefore, a number of these separations are discussed.

It is important in a report of this type to discuss the disadvantages as well as the virtues of the method, so a few pages have been devoted to the difficulties that occur. In some of the separations

it will be apparent that the technique is cumbersome if only because

of the length of time that the separation requires.

This author is indebted to all of the other authors of the

NAS-NRC Nuclear Science Monographs on the radiochemistry of the

elements, since frequent reference has been made to the detailed

distillation procedures listed in those reports.

II. THE DISTILLATION PROCESS FOR RADIOCHEMICAL SEPARATIONS

A. List of General References on Distillation

"Technique of Organic Chemistry," Volume IV. A. Weissberger, editor. Interscience Publishers Inc. New York (1950).

"Distillation Principles and Processes," Young. Macmillan Co. Ltd., London, 1922.

"Elements of Chemical Engineering," Badger and McCabe, 2nd edn. McGraw-Hill Publishers, Inc. New York, 1936. Chapter IX "Distillation."

B. Types of Apparatus

The distillation technique proves to be suitable for many radio-

chemical separations, since a large number of variables in the distilla-

tion process, e.g., pressure, temperature, time of distillation, vapor

condensing surface, etc., allows modification of the method to enhance

the separation.

In order to clarify the description of the radiochemical distilla-

tion separations which have been reported, the processes have been

grouped into two classes; the carrier vapor distillation train, and the

simple one plate distillation.

1. Carrier Vapor Distillation Train

All of the radiochemical methods in this class utilize an

apparatus that can be described as follows: The distillation flask

containing the solution to be separated is connected with a ground glass

joint through which is passed a tube for introducing reagents and/or a

carrier gas. The outlet tube which also passes through the joint is

terminated in a male connector of a ball and socket joint. The ball and

socket joint facilitates assembly of a multi-flask distillation train.

The vapor collecting flasks are made in a similar manner. The inlet tube is allowed to extend into the flask so that its tip extends below the collecting fluid. Any number of these flasks can be connected together to allow separation by selective condensation in each collecting flask through choice of the collecting medium. The vapor collecting flasks may be arranged to allow either heating or cooling as desired.

Quite often it is advisable to pass an inert gas through the distillation train to act as a carrier for the radioelement. It is best to draw the gas through the system so as to maintain the internal pressure slightly below that outside of the apparatus. This reduces leakage which could result in radioactive contamination.

Most often the train type of distillation requires the addition of usually ten milligram amounts of the naturally-occurring element to the distillation flask before distillation. This acts as an isotopic carrier for the radioactive isotope of the element to be separated.

An excellent general study on the relative volatilities from various types and compositions of acid solutions of many elements has been done by Hoffman and Lundell (22).

2. Simple One Plate Distillation

In its simplest form the apparatus consists of a furnace and a vapor collector. Figure 1 illustrates a particular system which uses the simple one plate distillation. The surface area of the collector is restricted to a few cm^2 so that the collector itself upon which the radioisotope is collected may be counted.

In order to maximize the yield of the distillation, the collector is placed as close to the furnace as is possible without causing excessive heat transfer through the radiative process. Such distillations are carried out in vacuum to reduce the dispersion of vapor and further optimize the yield. It should be realized that in vacuum the vaporization process is often a sublimation; however, for this general discussion the term distillation will be used. Stockendal (60)

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COLD FINGER 35 O.D.
TEFLON FILM
Cu WIRE RING
17 O.D.
VYCOR DEFLECTOR
6 O.D.
CARBON ROD FURNACE
VYCOR HOLDER
INDUCTION COILS
30
SAMPLE
DISTILLATION CHAMBER
PYREX CYLINDER 45 O.D.

FOR VACUUM GAGE
19/38
VACUUM RELEASE STOPCOCK 2 O.D.
80
165
35 O.D.
90 O.D.
FLANGE WIDTH 15 (GROUND FLAT)
COLD FINGER
90 O.D.
30
30 O.D.
95
45
36
40 O.D.
150
COLD TRAP
15 O.D.
TO VACUUM
120
48 O.D.

NOTE:
1. ALL GLASS TUBE STOCK
2. ALL DIMENSIONS IN mm

Fig. 1—Apparatus for vacuum distillation showing furnace with collector in place (left) and the complete apparatus showing location of furnace (right).

as well as the author (12) has found that a heated deflector in the shape of a cone directs the vapor toward the collecting device to give a more satisfactory yield.

Many types of furnaces have been described (10, 17, 59). The most common method of heating is with a spiral tungsten filament shaped to form a cone. Quite often a ribbon is used so that the sample to be heated can be placed on the ribbon. The filament is then heated by passing an electrical current through it.

Induction heating proves to be a successful method. It is particularly attractive when a vacuum apparatus is used since the induction coil need not enter into the vacuum. A carbon crucible-furnace heated by induction has been used for some separations, (12,21). Care must be taken with any furnace to ascertain that the radioisotopes to be distilled do not form a non-volatile compound with the furnace material. Carbides may be formed in a carbon furnace.

The choice of material which is used to collect the vapor is of great importance. It is possible to increase the selectivity of a separation by proper selection of the composition of the collector to improve the efficiency of collection of one over that of another radioelement. Even though considerable care is given to the formation of the surface of the collector it may be assumed that many gases are adsorbed on its surface even in a vacuum. The presence of an oxide film, adsorbed gases, or an otherwise unclean surface on the collector usually diminishes the collection efficiency. Good discussions of these collection problems are given in references (3, 8, 10, 15, 17, 18, 36, 43, 44, 54).

Thin plastic films are used as a collector, since this allows the deposited vapor to be readily removed for measurement of the radiation of the deposited radioisotopes. The back scattering of the radiation from a film of this type is minimized. Most of the film collectors are cooled by using a cooled backing surface such as the

outside of a liquid nitrogen cold trap upon which the film is fixed.
Merines (40) discusses the formation of homogeneous counting samples
by vacuum distillation, and Widmer and Kirsch (64) have evaporated
radioisotopes directly onto the surface of solid state radiation
detecting devices. Parker (45) describes an apparatus for the prepar-
ation of thin beta particle counting films. Holland (21) has prepared
a comprehensive text on the technique of vacuum deposition of thin
films. Yaffe (66) presents an excellent review of thin counting films.

Quite often a radioisotope which has been produced in a
nuclear reaction with a target material can be separated in a one step
process by simply heating the target material. Since the radioisotope
is free of a large quantity of the stable element (usually called
carrier-free), self-absorption of the radiation is minimized. Another
advantage is that the distillation technique provides a very even
distribution of the radioisotope on the collector.

Many of the designs for the distillation apparatus incorporate
a means for detecting the radiation from the separated radioisotope
soon after the deposition has ended (17, 45, 53). Such an apparatus
usually incorporates a shielded detector within the system so that the
collector does not have to be removed from the vacuum. With this
method, distillation proves to be a useful technique for measuring
radioisotopes with very short half lives (e.g., 1 second).

At present it has been found to be difficult to reproduce a
given yield in a carrier-free separation. This is probably due to
difficulty in the collection of the vapor. In addition, the yields
are often low. This can be due to lack of complete volatilization
as well as lack of quantitative vapor collection. Fortunately for
most applications, where the radioisotope is used for study of its
radiation, the yields have been sufficient. The high degree of purity
of the separated radioisotope makes the simple one plate distillation
method particularly suited to the carrier-free separation of products

from an irradiated target.

There has been much progress in the separation of macro amounts of substances from multicomponent systems. Satisfactory separations have been obtained through the use of a fractional sublimation technique which is analogous to the zone melting technique and has been called the vapor zone refining method (39, 55, 61, 63). The apparatus consists of a long cylindrical evacuated tube with the charge at one end. The area of the tube containing the charge is heated to sublime the volatile components. These components condense on the sides of the tube which are not heated. The heating element is then moved progressively down the tube resulting in a repetitive sublimation and increasing fractionation of the components. Another approach involves the repetitive evaporation and condensation of material on moving belts (7). Heat is applied from a stationary heating element over which a belt containing the substance to be sublimed is passed. A moving collector belt passes over a cooled portion above the heating element. Fractionation is accomplished by increasing the temperature and moving the collector and heating belts.

C. Expected Purity for the One Plate Distillation

Except in the simplest of cases it is very difficult to determine beforehand how satisfactory a separation by distillation will be. In the interest of indicating how the terms which are most often used in discussing radiochemical separations relate to terms of more common usage in the physical chemistry of the process, a few relationships will be given.

m_1, m_2 = moles of components 1 and 2 respectively in the pot, in the solid phase at any given moment during distillation.

n_1, n_2 = moles of components 1 and 2 respectively, in the condensate.

c_1, c_2 = moles of components 1 and 2 respectively, initially in the pot before distillation.

D = decontamination factor.

Y = fractional yield.

α = relative volatility.

If it is desired to separate the more volatile component 1 from component 2, using a single plate batch type of distillation (or sublimation), the decontamination factor from component 2 is often indicated in radiochemical separations to be $D = \dfrac{A_2^o}{A_2^1}$, while the yield for component 1 is $Y = \dfrac{A_1^1}{A_1^o}$, where A_2^o equals total radioactivity (counts per minute) of the contaminant present with the desired radioisotope before separation, A_2^1 equals total radioactivity (counts per minute) of the contaminant which separates with the desired radioisotope, A_1^o equals total radioactivity (counts per minute) of the desired radioisotope before separation, A_1^1 equals total radioactivity (counts per minute) of the desired radioisotope after separation. Since the specific activities of each component are not likely to be altered during the distillation (neglecting isotope effects),

$$D = \frac{S_2 A_2^o}{S_2 A_2^1} = \frac{c_2}{n_2} \qquad Y = \frac{S_1 A_1^1}{S_1 A_1^o} = \frac{n_1}{c_1}$$

where S_1, S_2 are the specific activities in disintegrations or counts per unit time per unit weight of the element or component.

The term "relative" volatility which is used to define the degree of separation expected is defined as $\alpha = \dfrac{\bar{P}_1 X_2}{\bar{P}_2 X_1}$, where $\bar{P}_1$, $\bar{P}_2$ are the partial pressures of components 1 and 2 which have mole fractions X_1 and X_2 in the distillation pot. When Dalton's law of partial pressures can be applied $\dfrac{\bar{P}_1}{\bar{P}_2} = \dfrac{z_1}{z_2}$, where z is the mole fraction in the vapor. Substituting into the equation for α gives

$$\alpha = \frac{z_1 X_2}{z_2 X_1} = \frac{n_1 m_2}{n_2 m_1}$$

If Raoult's law applies to the vapor, $\bar{P}_1 = X_1 P_1$, $\bar{P}_2 = X_2 P_2$ and $\alpha = \dfrac{P_1}{P_2}$ where P_1, P_2 are the vapor pressures of the pure components. Therefore α is a constant. At the beginning of a distillation where $n_1 \lll c_1$, $n_2 \lll c_2$, $\alpha = \dfrac{c_2 n_1}{c_1 n_2} = DY$. As the distillation progresses the product DY decreases according to the expression $\alpha' = DY = \dfrac{Y}{1-(1-Y)^{1/\alpha}}$.

Since a better separation is obtained from a distillation which has

a higher α' there is an optimum yield which must depend upon the purposes of the separation. If a high purity is desired the yield must be sacrificed.

The above is a considerable oversimplification of a practical distillation since the formation of non-ideal solutions and non-ideality of the vapor can greatly alter the separation from that indicated above. Even less can be predicted about the separations obtained with carrier-free systems than with macro systems. Nevertheless, one might be inclined to be optimistic about the uses of the method when one considers that vapor pressures of many substances, e.g. metals, differ in magnitude by many powers of ten.

D. Future Applications.

Even though new methods are found each year for the separation of compounds from aqueous solution by distillation, it appears that one of the most promising volatilization techniques is the vacuum distillation of the compounds or elements. This is particularly true if carrier-free separations are necessary, because wet chemical methods involve necessarily the addition of chemical reagents to the system. Additions of such chemicals add trace quantities of the inactive elements. Therfore, it is advantageous to have a separation system which requires contact of the carrier-free element with a minimum number of other elements. A simple one plate distillation with a furnace and one vapor collector in an evacuated system affords less carrier contamination of the separated radio-isotope than most other separation techniques, except possibly the mass spectrographic method.

Additional emphasis could be placed on the use of compounds such as the solid chlorides in order to enhance selectivity of a particular separation.

Since the carrier-free vacuum distillation technique can often be carried out very rapidly, its use for measuring the properties of short lived radionuclides should be given greater attention.

III. RADIOCHEMICAL DISTILLATION PROCEDURES.

This section is a compilation of radiochemical separation procedures using distillation techniques. No attempt has been made to indicate in detail all of the distillation procedures which have been used for the various elements. If the element has been omitted no use of distillation technique was found in the literature. Selection of the included procedure has been based upon the author's evaluation of the description in the literature with particular regard to the supposed ease of carrying out the procedure. A number of the methods described here have been attempted by the author, and comments are given with these methods. Experimentalists who feel that their procedures should be included are invited to write to the author so that the method may be considered for a later edition.

Many of the distillation procedures for the elements have been included in the other National Academy of Sciences monographs dealing with radiochemistry of the individual elements. In order to reduce repetition, reference is made to these procedures in this monograph and it is suggested that the reader consult the pertinent monograph concerning details of the distillation procedure for the element of interest.

A. _Hydrogen._ These separations usually involve the conversion of tritiated compounds into tritium. The tritium is then passed into a counter for measurement of its radioactivity. Many of the techniques are reported in experiments done by geochemists who are interested in the tritium content of terrestial matter.

B. _Rare Gases._ He, Ne, Ar, Kr, Xe, and Rn - Most of the separation procedures involve passing a carrier gas through a solution of the rare gases at room temperature. The rare gas is then collected on a cold trap or adsorbed on charcoal. This technique provides a very rapid separation as described in Monograph NAS-NS 3104 (30). A most comprehensive compilation of methods for the separation of the rare gases by

distillation is given in Monograph NAS-NS 3025 (41).

C. Carbon, Nitrogen, and Oxygen. One of the most common methods for
purifying these elements in organic compounds is the distillation
technique. Since the number of applications far exceeds the scope of
this monograph no attempt will be made to review this work. C^{14} can be
counted with almost 100% efficiency by converting the C^{14} into a
molecule that is used as a counting gas. C^{14} containing methane,
acetylene and carbon dioxide have been successfully used as counting
gases. The most familiar C^{14} dating technique utilizes such procedures.

Nitrogen compounds are usually converted to NH_3 for separation by
distillation. A few specialized distillation procedures are detailed in
Monograph NAS-NS 3019 (23), and NAS-NS 3104 (30).

D. Halogens. Chlorine, bromine, and iodine are often separated by
the use of distillation. Distillation is used successfully for many
short-lived radioisotopes of these elements. (See Monographs NAS-NS 3005
(29), NAS-NS 3104 (30)). The usual procedure is to render the halogens
non-volatile by oxidizing them to the +5 valence state. The more
volatile components are then distilled and subsequently the halogen is
reduced to the element for distillation. Quite often a carrier gas such
as air or nitrogen is passed through the system to sweep out the halogen.

E. Alkali Metals. Na, K, Rb, Cs, Fr. The handling of the elemental
state of the alkali metals is difficult because of their reactivity
with O_2 and moisture. Most of the compounds of these elements are non-
volatile compared with the majority of those of the other elements.
However, techniques have been used for the volatilization of francium
which has been separated by carrying it on cesium silicotungstate,
Monograph NAS-NS 3003 (24). The precipitate is converted to the per-
chlorate and the cesium and francium are removed as the elements by
means of a "flash distillation," in which the precipitate on a platinum
plate is touched with a gas-oxygen flame for a fraction of a second.
The cesium and francium are collected on another platinum plate which

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is placed a few millimeters away. The platinum collector plate is then measured for radioactivity.

F.   Silicon. The distillation procedures for silicon all utilize the fact that SiF_4 is volatile (boiling point at 760mm = -65°C). Several detailed procedures are indicated in Monograph NAS-NS 3049 (35).

G.   Phosphorous. A carrier-free distillation technique which was used successfully for Sr^{90} is described by Sherwin (53, 54) for P^{32}. (See I below).

H.   Sulfur. The usual techniques in which the sulfur is converted to SO_2 or H_2S are applicable in a radiochemical separation. No detailed procedures were found in the literature.

I.   Alkaline Earths, Rare Earths and Yttrium. The only description of a separation by distillation is given by Sherwin (53) where carrier-free Sr^{90} (28y) is separated from Y^{90} (64h) by a technique which can best be described as a simple one plate vacuum distillation. The sample is heated on a tungsten filament. Both the collector and the filament can be adjusted from outside the evacuated chamber. The vapor is collected and measured with a Geiger counter while still in the apparatus. Since adequate shielding of the counter from the sample is provided in the evacuated chamber the system allows the measurement of short lived radioisotopes. The separation results in a decontamination factor of 10^4 for Y^{90} from Sr^{90}.

J.   Vanadium. A procedure is described in Monograph NAS-NS 3022 (6) where the volatile VCl_4 and $TiCl_4$ are distilled together from scandium chloride.

K.   Chromium. A standard procedure for the distillation of chromium uses chromyl chloride (CrO_2Cl_2). This radiochemical separation is done by predistilling potential contaminants such as Sn, Sb, As and V as their volatile halides. DeSoete reports the success of this procedure in Monograph NAS-NS 3007 (49). The contaminants are Sn (0.03%) and V (0.02%).

L.   Manganese. A unique method for the distillation of manganese
utilizes the volatility of permanganic acid (48). The apparatus is
similar to the distillation train described in Chapter II.

The Isolation of Radiomanganese by Distillation as Permanganic Acid (48)

Procedure:

Introduce the aqueous solution into the distillation flask. Add
H_2SO_4 (36N) to make 10 M in sulfuric acid. If chemical yield is to be
determined, add 0.5 mg of manganese carrier. Add 1 gm KIO_4 and heat the
mixture using an iso-mantle or a metal bath. Bubble an inert gas through
the solution to ensure regular boiling and keep the volume constant by
the addition of 40% HNO_3, saturated with KIO_4. Distill 10 minutes.
Boil the distillate with 250 mg KIO_4 and dilute to 25 ml for spectro-
photometric determination of yield. (See Sandell, E. B., "Colorimetric
Determinations of Traces of Metals," Interscience Publishers, Inc.,
New York, 1959, p. 608).

Remarks:

Under the given conditions Fe, Co, Cr do not interfere. The pro-
cedure was tested with manganese concentrations ranging from carrier-free
to 1 mg. The method was successfully used for the carrier-free separa-
tion of manganese from deuteron-irradiated iron cyclotron targets and
gave also reproducible results in activation analysis of manganese in
various biological materials. The recoveries were 60 to 90%.

M.   Germanium, Arsenic, Antimony, and Tin. These elements are usually
separated as a halide or as the hydride. Selected separation procedures
are given in Monograph NAS-NS 3002 (4), NAS-NS 3033 (37), NAS-NS 3043
(38), NAS-NS 3023 (42) and (62). A history of the analytical separation
of As, Sb, Sn by distillation of the halides is given by Scherrer (51).

An interesting analog of the zone melting technique is the "vapor
zone refining" method described by W isberg and Rossi (63). This
method involves heating a tube of arsenic under vacuum. The heating
element is slowly moved along the tube in a manner similar to that

employed in zone melting. In this way impurities Se, Fe, Al and Cu are concentrated by a factor of three.

Ge^{73} (half-life 0.33 second) has been separated from As^{73} by a "flash distillation" technique (14). This technique uses the basic idea of the single plate distillation, but the source is heated rapidly to its melting point with a large current surge. The distillation is completed in a fraction of a second, thereby allowing measurement of the short lived Ge^{73} which vaporizes and collects on a condenser plate situated a few millimeters from the source.

Successive Separation of Arsenic, Antimony and Tin (9)

Procedure:

The degree of separation of one milligram amount of each of the elements is measured by the use of radioisotopic tracers.

1. The final concentration of the solution after dissolving sample should be 11 N HCl.

2. The solution is then distilled in an all glass apparatus (e.g. similar to the distillation train described in Chapter II.) at 109°C while drawing anhydrous CO_2 through the system. The vapor is collected in 11 N HCl (Note 1).

3. Without interrupting the heating add 7 ml of concentrated H_3PO_4 (85) (Note 2) to the distillation flask. Heat this solution to 160°C and then slowly add 150 ml. of 11 N HCl while maintaining the temperature between 155 - 165°C (Note 3).

4. Lower the temperature to 140°C and add a solution of 3 parts 11 N HCl to 1 part 40% HBr by slow addition to the distillation flask so that the temperature is maintained at 140°C (Note 4).

Notes: 1.   Quantitative separation of arsenic requires 40 min.

2.   The tin complexes with phosphate and does not distill.

3.   Quantitative separation of antimony is accomplished in 110 min.

4. Quantitative separation of tin is accomplished in 75 min.

5. The chloride fractions can then be counted in solution or precipitated (e.g. as oxide, sulfide, etc.) and then counted.

N.   Selenium and Tellurium. Selenium is routinely separated by distillation of the bromide using the wet-chemical distillation train method described in Chapter II. Tellurium bromide is volatile also, but careful distillation in the appropriate solution can separate the two. Most of the other volatile bromides and chlorides contaminate the separation. Detailed separation procedures are given in Monograph NAS-NS 3030 (32), NAS-NS 3038 (31).

Carrier-Free Isolation of Selenium from Arsenic

Procedure:

An interesting carrier-free procedure devised by Garrison, et al (19) separates the carrier-free radioisotope of selenium formed by the reaction As^{75} (d, 2 n) Se^{75}.

1. Dissolve the arsenic metal in a minimum volume of aqua regia.

2. Add 12 N HCl to remove the excess nitric acid and adjust the acidity to 3 N HCl.

3. Add 10 mg of tellurous acid (H_2TeO_3) and precipitate tellurium metal with SO_2. (95% of the Se^{75} is carried on this precipitate).

4. Dissolve the tellurium metal in 16 N HNO_3 and add arsenic chloride.

5. Reprecipitate the tellurium metal and filter to remove radio-isotopes of arsenic produced by side reactions such as As^{75} (d,p) As^{76}.

6. Repeat steps 4 and 5 two more times.

7. Dissolve the tellurium metal in the minimum volume of 16 N HNO_3.

8. Distill in an all glass apparatus at 200°C (similar to train distillation described in Chapter II), after adding 14% HBr using nitrogen gas carrier. The carrier-free selenium distills as the tetra-bromide while the tellurium remains.

O.　　Ruthenium, Osmium, Technetium, Rhenium.　These elements have been

grouped together because their separation by distillation is done by

volatilizing the oxides, RuO_4, OsO_4, Tc_2O_7 and Re_2O_7.　Selectivity in

the separation can be affected by varying the composition of the

distillation mixture and the composition of the solutions in which the

vapor is collected.

Detailed separation techniques are given in the Monographs for

ruthenium NAS-NS 3029 (65), osmium NAS-NS 3046 (34), technetium NAS-NS

3021 (1), and rhenium NAS-NS 3028 (33).

P.　　Rhodium and Silver.　A carrier-free technique is described by

Parker (47) for the separation of Rh^{103} from a ruthenium target.　Yields

of 20% were obtained in a one-minute distillation.　The apparatus is

similar to that using the simple one plate vacuum distillation technique

described in Chapter II.　The ruthenium target is placed on a tungsten

filament, heated and rhodium is collected on Zapon film.

For silver, no aqueous solution techniques were found in the

literature but several methods used distillation to separate the

carrier-free radioisotope from a target material.　The simple one plate

distillation technique described in Chapter II was used in these

procedures.　The carrier-free technique used by Parker (47) for Rh^{103}

was also used to separate Ag^{105} from a rhodium target.

Procedure 1.

Separation of Silver from Palladium (12)

Reaction: Pd^{108} (n,γ) Pd^{109m} (13.5h)　$\xrightarrow{\beta-}$ Ag^{109m} (39s).
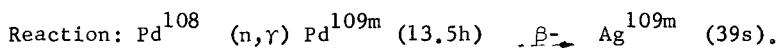
The palladium foil is placed in a carbon furnace and heated by

induction for three minutes at the temperatures indicated in the table

below.　The collected vapor which was invisible was placed in a

scintillation well counter.　The 39 second half-life of Ag^{109m} was

detected.　In a short time the Ag^{109m} reached secular equilibrium with

the residual parent Pd^{109} which had volatilized with the carrier-free

silver.　This allowed determination of the Pd^{109} contamination in the

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Ag^{109m} as follows:

Temperature °C	Yield %	%Contamination of Pd^{109} in Ag^{109m}	Decontamination Factor for Pd^{109}
780	0.07	11	1.4×10^4
880	0.14	3	2.3×10^4
980	1	0.18	5.2×10^3

Procedure 2.

Separation of Silver from Rhodium (57)

Reaction : Rh^{103} (α,n) Ag^{106}
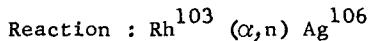
1. Heat the rhodium metal target on a tantalum filament to approximately 2000°C in vacuum (near the m.p. of rhodium) (Notes 1 and 2).

2. Collect the vaporized Ag on a cylindrical quartz catcher. (Note 3).

3. Dissolve the deposit in a few drops of concentrated HNO_3 and one drop of concentrated HF. (Note 4).

4. Evaporate carefully to dryness and dissolve into 2 M HCl.

5. Pass in O_2 to oxidize any Pd to the +4 state.

6. Pass through a column (3mm dia x 2 cm) of Dowex 1.

7. Dilute Ag with 10 M HCl.

8. Dilute to 2 M HCl and repeat the ion exchange step.

9. Evaporate to dryness, dissolve into a solution which contains 2.5 ml concentrated nitric acid, 2.5 ml concentrated sulfuric acid and 0.5 gm of urea per 100 ml of solution.

10. Electrolyze for two hours at 6 milliamperes current onto 0.010 dia. platinum wire. (Note 5).

Notes: 1. When heated for only a few seconds almost 100% volatilization of Ag is obtained.

2. The tantalum filament is a 1/4" wide x 0.15" thick strip.

3. Some rhodium metal deposit was visible.

4. The rhodium metal did not dissolve appreciably.

5. The overall yield was 50%.

Q. Cadmium

Procedure 1.

Electrolysis of Cadmium into Mercury Electrode and Distillation of

Residue (12).

One milligram of cadmium is electrolyzed into a mercury cathode.
The macro mercury is removed and the cadmium is distilled and collected.

The following procedure was used:

1. Place 1/2 gm. of mercury metal into the mercury cathode cell.
(Note 1).

2. Place 1 mg. of cadmium as a carrier solution in the cell with
trace amounts of the radioactive contaminating element (or with radio-
active cadmium tracer added if yields are to be determined).

3. Add sufficient 1 N H_2SO_4 to bring the total volume to approx-
imately 1 ml.

4. Electrolyze the solution for 30 minutes at 6 volts.

5. Remove the electrolyte by means of a suction tube, and discard.
Wash the mercury drop five times in distilled water and three times in
acetone, and transfer to a pyrex tube. Evaporate the mercury. Transfer
the residue to a carbon rod furnace and distill the cadmium between
160° and 180°C at about one micron of pressure until no visible deposit
forms when a fresh collecting surface is used after successive minute
distillations.

6. The area on the Teflon film which contains the deposit is placed
in a counting tube and counted in a scintillation well counter.

Notes: 1. Make a simple mercury cathode electrolysis cell by sealing
           a platinum wire into the bottom of a 5 ml. Pyrex test tube.

        2. An experiment in order to evaluate the general procedure
           was done by attempting the separation of cadmium from the
           fission products of uranium. Uranyl acetate (0.100 gm) was
           irradiated in the Phoenix reactor of the University of

Michigan for 10 minutes at 10^{12} neutrons per cm^2 per sec.

Two hours after irradiation the solution was dissolved in

1 N H_2SO_4 containing 1 mg. of cadmium carrier, and was

electrolyzed for 30 minutes. This procedure removed 97%

of the fission product activity from solution. After

separating the macromercury and distilling the cadmium at

180°C in the vacuum distillation apparatus, detectable

amounts of Cd^{115} and Cd^{117m} were found. (1000 c/m in the

photopeak using a 3" x 3" potassium iodide scintillator

with a gamma ray spectrometer). Estimation of its purity

indicated that the decontamination factor for the total

activity in the fission products at 3 hours after fission

was 10^5.

3. The apparatus which was used is shown in Figure 1. A

dessicator type of ground glass seal allows easy access to

the distillation chamber. The furnace consists of a hollow

carbon rod which is heated externally with induction coils.

The Teflon film is wrapped around the cold finger.

4. Since 1 mg. of cadmium is used as a carrier, self

absorption corrections will be an important consideration

when doing absolute assay of the radioisotope.

Data from a detailed investigation of the separation are given in Table I.

Procedure 2.

Electrolysis of Cadmium into Copper Foil and Distillation from the Foil.

This procedure is given in Mongraph NAS-NS 3001 (11).

Procedure 3.

Separation of Cadmium from Silver (25).

Reaction: Ag^{107} (p, 3n) Cd^{105} (55m)
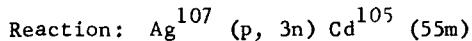
The silver target is heated in an evacuated quartz furnace to its

melting point (about 960°C). The carrier-free Cd^{105} is collected on a

TABLE I

DECONTAMINATION FACTORS FOR ELECTROLYSIS OF CADMIUM INTO MERCURY
AND THE DISTILLATION OF CADMIUM

Contaminant	Amount (μg)	Decontamination Factor Electrolysis	Distillation	Overall
Ag^{110}	8	2	10	20
$Ce-Pr^{144}$	C.F.**	1.4×10^3	20	3×10^4
Cs^{134}	3	10^3	20	2×10^4
Co^{60}	7	2.5	10^3	2.5×10^3
Ir^{192}	.05	4	2.3×10^2	10^3
Se^{75}	400	26	1.6	42
Ru^{106}	3.6*	7	10^3	7×10^3
Tl^{204}	580	1.7	4.1	7
Zn^{65}	50	1.6	20	32
Zr	0.8	3	27	81

*Solution of chloride converted to sulfate so that final concentration is 0.1 N H_2SO_4.

**C. F. = Carrier-Free.

Cd yield (1 mg.) 93 ± 2.5% error is standard deviation, 5 experiments.

1/8" dia. aluminum disc (1.7 mg/cm^2) or a tungsten wire 3/4" long and 10 mils in diameter. The degree of separation or yield was not discussed.

Procedure 4.

Separation of Cadmium from Silver (16)

Reaction: Ag^{107} (p,n) Cd^{107} (6.7h) $\xrightarrow{\beta+}$ Ag^{107m} (44s) (16)
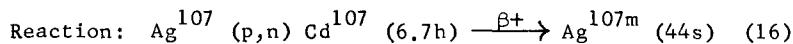
The silver target is heated on a tungsten filament, and the cadmium separated at 1400°C at a pressure of 10^{-5} mm Hg. The vapor is collected on silver (metal). Only 0.5% of Cd^{107} was collected, but the collection efficiency for Cd^{107} was found to increase to 50% when the silver surface was freshly prepared in vacuum such that surface absorption of gases was minimized.

R. Indium

Separation of Indium from Tin (12)

Procedure:

Reaction: Sn^{112} (n,γ) $Sn^{113}_{(112d)}$ $\xrightarrow{K}$ In^{113m} (1.7h) (12)
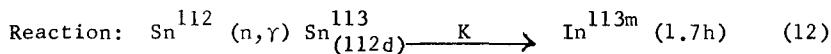
The tin metal in granular form was heated in a carbon furnace by induction in a vacuum of 5×10^{-4} mm Hg for 20 minutes at 270°C. The In vapor was collected on Teflon 0.0001 inches thick stretched over a glass cold finger at liquid nitrogen temperature. (See Figure 1). A yield of 4.5% was obtained. When 0.001" thick tin foil is used, an In^{113m} yield of 52% is obtained in 20 minutes.

S.   Gold. These methods utilize the single-plate distillation technique described in Chapter II. Au^{197} can be produced from platinum metal by the reaction Pt^{198} (n,γ) $Pt^{199}_{(30m)}$ $\xrightarrow{\beta-}$ $Au^{199}_{(3.2d)}$ (28). The platinum metal is heated almost to its melting point and the gold is almost completely volatilized. No information was given on the type of collector used.

Another application by Parker (47) volatilizes Au^{197} and Au^{195} from a platinum target. About 20% yield was obtained in a one-minute heating (no temperatures are indicated).

T.   Mercury. Mercury is often distilled as a metal which has been obtained after a reduction step to gain added decontamination. Mercurous chloride has also been used in a distillation separation. Several of these procedures are discussed in Monographs NAS-NS 3104 (30), NAS-NS 3026 (50).

Procedure 1.

Steam-Distillation of Mercury (56).

Sion and Hoste have stated (56): "Steam distillation of mercury in the presence of a reducing agent was first proposed by Miller (W. L. Miller & L. E. Wachter: Anal. Chem. 22, 1312, 1950), who claimed quantitative recoveries. In our procedure the reducing agent (either $SnSO_4$ or $SnCl_2$) was added in one step instead of dropwise. A micro-Kjeldahl apparatus was used. The distillation process was monitored with Hg^{197} - Hg^{203} tracer.

Procedure:

Introduce the solution into the micro-Kjeldahl apparatus and add

H_2SO_4 to make the solution 0.5 N in acid. The total volume should not exceed 20 ml. Add 20ml. $SnSO_4$ or $SnCl_2$ solution. Steam distillation is carried out from 15 to 20 minutes, the tip of the cooler plunging into 25 ml. of a H_2SO_4 - $KMnO_4$ solution, cooled in ice water.

Reagents:

1. Conc. H_2SO_4 analytical grade.

2. Reducing agent:

    a) 10 g $SnSO_4$ dissolved in 100 ml. H_2O + 1 dr 36 N H_2SO_4; filter off residue;

    b) 125 g $SnCl_2 \cdot 2H_2O$ are dissolved in 100 ml of 12 N HCl and diluted with water to 1000 ml;

3. H_2SO_4 - $KMnO_4$ mixture: 250 ml 3.5 N H_2SO_4 + 250 ml 0.8 N $KMnO_4$.

Remarks:

Method was tested with Hg carrier in the 0.2 µg range. Quantitative results were obtained for up to 78 µg.

Alternative procedure:

Distillate is collected in two one ml portions of the H_2SO_4 mixture in 2 consecutive micro-traps."

Procedure 2.

Separation of Mercury Deposit from Metallic Copper (12)

This method uses the well-known fact that copper metal will reduce mercury onto its surface from aqueous solution. The copper strip is then heated to remove the mercury.

1. To a 50 ml round bottom centrifuge cone, add carrier-free Hg^{203} tracer solution, in known amount for yield determination. If a decontamination factor is to be measured, use inactive mercury plus a radioisotopic tracer of the contaminant.

2. Add sufficient acid to bring total volume to 2 ml. of 0.15 N HNO_3.

3. Prepare an 8 mg. Cu foil, 0.5 cm^2 (10 mils thick), by washing in 1 N HNO_3 to clean the surface; rinse with distilled water and add to the centrifuge cone.

4. Stir slowly for about 20 minutes and wash six times with
distilled water and once with acetone.

5. Transfer to a carbon rod furnace, and heat to 100°C for three
minutes.

TABLE II

DECONTAMINATION FACTORS FOR THE SEPARATION OF MERCURY ONTO
COPPER AND THE DISTILLATION OF MERCURY

		Decontamination Factors		
Contaminant	Amount(μg)	Chemical Redn.	Distillation	Total
Ag^{110}	200	1.2	2×10^4	2.4×10^4
Ba^{140}	C.F.	$3 \times 10^2 (2.3 \times 10^5)$	3.3×10^2	$10^5 (7.6 \times 10^7)$*5
Au^{198}	220	40	4×10^3	1.6×10^5*2
$Ce^{144}-Pr^{144}$	C.F.	1.2×10^3	2.3	2.8×10^3
Cd^{115m}	190	1.6×10^2	3.4 *3	5.4×10^2
Co^{60}	7.4	10^5	5	5×10^5
Cr^{51}	5	2.5×10^3	6	1.5×10^4
Cs^{134}	2.5	3×10^3	2.3	6.9×10^3
Cu^{64}	65	1.2×10^2	1.4×10^2	1.7×10^4
In^{114}	4.6	$10^5 (2 \times 10^7)$*4	10^2	$10^7 (2 \times 10^9)$
Ir^{192}	0.2	2×10^4	1.4	2.8×10^4
Nb^{95}	8	3×10^3	10	3×10^4
Pd^{109}	50	1.4	1.7×10^5	2.4×10^5
Ru^{106}	2	32	350	1.1×10^4
Se^{75}	3.5	6.5	2.8×10^3	1.8×10^4
Sb^{124}	0.6	2.8×10^2	4.7	1.3×10^4
Sn^{113}	500	1.3×10^3	10	1.3×10^4
Ta^{182}	4	-	-	--*1
Tl^{204}	400	4×10^4	2.5	10^5
Zn^{65}	180	4.3×10^3	4.7	2×10^4
Zr^{95}	8	2.7×10^3	14	3.8×10^4

*Notes (Table II):
1. Fluoride complexing agent for Ta interferes with the mercury
   yield.
2. Mercury comes off at 350°C.
3. Mercury comes off at 220°C.
4. Start with 5×10^6 and (4×10^8) (C/M resp.)
5. Start with 1.9×10^6 and (1.9×10^9) (C/M resp.)

6. Collect the mercury on Teflon film. Cut out the area where collection occurs, and count in a scintillation well counter.

Notes: 1. The yield of the separation with 76 μgm of Hg in 0.15 N HNO_3 is 72 ± 3%. Error is standard deviation for 5 experiments.

2. Decontamination factors for a number of elements are listed in Table II.

Procedure 3.

Separation of Mercury from Gold (5)

Reaction: Au^{197} (n,γ) Au^{198} $\xrightarrow{\beta-}$ Hg^{198} (n,γ) Hg^{199} (42m)

Neutron-irradiated gold foil is heated in a vacuum at 10^{-5} mm Hg at 900°C (quartz oven) for 5 to 6 hours. The Hg vapor is collected in a U-tube under liquid nitrogen. A 90% yield is obtained.

Procedure 4.

Separation of Mercury from Gold (46)

This technique uses the one-plate distillation described in Chapter II with an apparatus very similar to that shown in Figure 1. Mylar is wrapped around a cold finger to act as a collecting surface. The charge is heated by a flame in a quartz tube under vacuum.

U. Lead. Most of the distillation techniques are used for the separation of a rare gas precursor. Then the lead is separated by chemical methods from other daughter products of the rare gas radioisotope. (62).

Carrier-Free Separation of Pb^{212} (ThB) from Parent Th^{232} (27)

Procedure:

1. Place Dowex 50 (8x-100-200 mesh) cation exchange resin in a Soxhlet extractor. Prepare the Dowex 50 by washing in 6 N HCl.

2. Reflux thorium nitrate in 1 M HNO_3. Notes 1 and 2.

3. Remove the ion exchange column (22 x 55 mm) from the extractor and dilute the Pb^{212} with 1 N HCl.

Notes: 1. Several nitric acid concentrations were tried. In 1 M
HNO$_3$ a 40% yield is obtained.

2. Presumably Rn^{220} (55 sec) is volatilized and the daughter
Pb^{212} is selectively retained on the ion exchange column.

V. <u>Bismuth.</u> Distillation is used mainly, as in the case of lead, to separate parent rare gas radioisotopes. (62).

W. <u>Polonium.</u> A complete description of the distillation techniques used for polonium are given in Monograph NAS-NS 3037 (13) and reference (61). The metal and various compounds such as the sulfide, diphenylcarbazide, and the diphenylthiocarbazide are volatile. Several of these have been used in separation procedures indicated in Monograph NAS-NS 3037 (13).

A technique for purifying polonium (Po^{211}) is to volatilize At^{211} from platinum while retaining Po^{211} (0.5 sec). (58).

X. <u>Astatine.</u> Since the chemistry of this element is quite complex the distillation technique has been used with more favorable results than those obtained with chemical methods in many separations. A very elaborate procedure is given in Monograph NAS-NS 3012 (2). Many of the older techniques are listed in (62). Others are listed in Monograph NAS-NS 3104 (30). A good discussion of the distillation technique for purification of astatine is given by Johnson et.al. (26).

Y. <u>Uranium.</u> Volatilization of uranium as UF$_6$ is a well-known separation procedure. Detailed procedures, particular studies on the decontamination factors, and interference effects are discussed in Monograph NAS-NS 3050 (20) and reference 52.

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