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A STUDY OF TECHNIQUES FOR REACTOR EFFLUENT ANALYSIS

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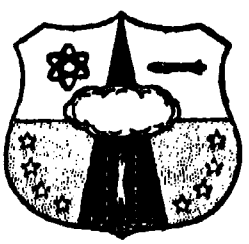

Research Development
AIR FORCE SPECIAL WEAPONS CENTER
Air Force Systems Command
Kirtland Air Force Base
New Mexico

Project No. 1448

(Prepared under Contract AF 29(601)-2852 by Rodney Melgard of Tracerlab, Incorporated, Reactor Monitoring Center, 2030 Wright Avenue, Richmond 3, California)

HEADQUARTERS
AIR FORCE SPECIAL WEAPONS CENTER
Air Force Systems Command
Kirtland Air Force Base
New Mexico

ABSTRACT

Methods for quantitative analyses of effluent samples from direct cycle air-cooled nuclear reactors are presented. Methods have been developed for analyses of beryllium and uranium at 10^{-9} gram levels in various collection media, and the level of beryllium and uranium present in the unexposed media has been determined. Separation and decontamination schemes have been developed for the detection of the following radionuclides: Mo^{99}, $Sr^{89,90}$, $Ru^{103,106}$, $Cs^{136,137}$, Ba^{140}, Y^{91}, Cd^{115} and Np^{239}. Collection media investigated included several filter papers, canisters of activated carbon, and gummed paper.

PUBLICATION REVIEW

This report has been reviewed and is approved.

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A STUDY OF TECHNIQUES FOR REACTOR EFFLUENT ANALYSIS

A. INTRODUCTION

The following report is the final report of the radiochemical and analytical development work which was performed by personnel of Tracerlab, Incorporated during the period June 15 - December 15, 1960, in support of Contract AF 29(601)-2852, and satisfies the requirements of Part II, paragraph A.2 of the above contract. This contract, which is under the technical direction of the Air Force Special Weapons Center, provides for the development of methods and techniques for the quantitative analysis of reactor effluent for beryllium, uranium and selected fission products.

The basic problem involved the establishment of techniques capable of defining the chemical and physical nature of effluent released by direct cycle ceramic core reactor fuel elements. Inasmuch as beryllium and uranium may be released in sensible quantities and are significantly toxic, the refinement of procedures capable of detecting these elements on the 10^{-9} gram level with 10 percent accuracy was required. Also, to detect the release of any significant quantity of the fission product inventory, a separation and decontamination scheme was required for the detection of any three of the following radionuclides: Mo^{99}, $Sr^{89,90}$, $Ru^{103,106}$, $Cs^{136,137}$, Ba^{140}, Y^{91}, Cd^{115} and Np^{239}. Three percent statistical accuracy was required.

As required in the contract, the level of beryllium and uranium present in the sample collection media was determined. Also, the beryllium, uranium and long-lived radionuclide content of the ground level atmosphere at the Nevada Test Site was determined. A method was required for the correction of background data obtained from radiochemical analysis of exposed filters for the incorporated background from the air, dust and filters.

As an aid to determining the various aspects of the reactor operations and in separating true effluent uranium from background uranium, the contract required the investigation of mass spectrometric analytical techniques for determining the isotopic composition of emitted uranium.

Also required as a reactor diagnostic tool was a feasibility investigation of applying particle study techniques for determining the relationships between emitted particle size and physical properties to particle radiochemical composition.

The following discussion emphasizes broad radiochemical and analytical problems encountered during the developmental work and also covers the general scheme of approach to the specific projects outlined above and their interrelationships.

The body of this report discusses the specific techniques developed by Tracerlab personnel during the contract period. It also includes the results of the many background determinations performed at the Richmond and Waltham laboratories.

The finalized procedures for the dissolution of the various collection media, separation and determination of beryllium, uranium, the radionuclides, and particles, as well as counting techniques where applicable, are included in the appendix.

B. DISCUSSION

In support of Contract AF 29(601)-2852, Rodney Melgard, Project Leader and Leon Leventhal, Manager, Technical Services, of the Reactor Monitoring Center met with Lt. Col. I. J. Russell of the Air Research and Development Command on June 23, 1960, at AFSWC Headquarters, Kirtland Air Force Base. The purpose of this meeting was to discuss in detail the work statement of the contract and review pertinent information available to Lt. Col. Russell to assist in performance of the contract. As a result of this meeting the timing and direction of effort were determined.

Discussion on the limits of detection and error limits indicated that uranium could be detected readily within the limits specified but that beryllium would be more difficult.

At this meeting it was also decided that an exposed background blank for uranium and beryllium would be necessary before actual implementation of the program.

On June 30, 1960, Mr. Leventhal met with Dr. David Bandel, Assistant Technical Director for Tracerlab's Eastern Division Laboratories in Waltham, Massachusetts. They discussed in detail, the techniques of emission spectrographic detection and determination of beryllium. It was agreed that TLW would supply Waltham with ashed filters for quantitative beryllium analysis by this method. Dr. Bandel will furnish the results and a procedure for this analysis.

On invitation from AFSWC, Rodney Melgard visited the Nevada Test Site on September 26 - 27, 1960. The purpose of this trip was to view the on-site monitoring equipment and nonradioactive tracer studies being carried out by Convair, Fort Worth, personnel. In addition, Mr. Melgard met with Lt. Robert Jacobs of AFSWC and discussed progress of the contract. This visit assisted in obtaining a better knowledge of the problems associated with the actual depositions which might be found on the filtering media.

Filters were received from R. L. Seale, Chief of Nuclear Design and Operations, of Convair, Fort Worth, Texas on August 9, 1960. These included nine 1106B Hi-volume fiberglass filters, five pleated filters, ten Millipore membrane filters, six activated charcoal filters and six carbon cartridges. Also received were ten 1106B filters, four pleated filters, six Millipore membrane filters and ten sheets of sticky paper from Lt. Col. R. E. Heft, of McClellan Air Force Base, Sacramento, California.

Information was requested from the Mine Safety Appliance Company, the Staplex Company and the Millipore Filter Corporation regarding composition of these filters, dissolution techniques, beryllium and uranium blanks, total weight and ash content. No pertinent data was obtained from any of these sources although various procedures were obtained outlining sample collection procedures for gross alpha, beta and gamma determinations. However, a quote from a letter received from R. B. Evans, Product Line Manager, of the Mine Safety Appliance Company, is interesting in view of the anticipated future use of their filters for effluent sampling. The quote is as follows:

> I think it would be impractical to use any of the Ultra-Aire Space Filters as sampling filters because of the potentially high blanks which could vary greatly from filter to filter. The rubber base adhesive and the corrugated asbestos board, as well as the filter medium itself, cannot properly be controlled for use in critical sampling techniques.
>
> The pleated hi-volume filters are manufactured in Pittsburgh - - - - from glass microfibers and natural esparto glass and are processed using unfiltered Pittsburgh drinking water. Again, they are not manufactured as analytical grade filters.

Six 1106B (fiberglass) filters, each of which had been exposed at the Nevada Test Site for approximately five hours, were received for determination of blank uranium and beryllium. In addition, radionuclide blanks were determined. Detailed results are tabulated later in the report.

Several factors governed the direction of development of techniques for detecting the various pertinent components of the reactor effluent. The dominating problem concerned the level of natural uranium and beryllium background found in all sample collection media and also in the chemical reagents used in subsequent isolation of the desired components. This concern necessitated the use of a carrier-free dissolution technique utilizing a minimum of highly purified reagents. Subsequent aliquotting of the carrier-free solution for the detection of beryllium and uranium kept these determinations isolated from the major portion of carriers, tracers, and chemical reagents and glassware which would be likely to add unknown amounts of natural contaminants.

Another consideration affecting the development of dissolution methods for the proposed sample collection media was the inclusion of ruthenium, molybdenum and cesium in the fission product assay. These elements tend to volatilize if heated above 500° C. Thus, an upper limit of 450° C was established for procedures enveloping ashing techniques for the destruction of the carbonaceous material in the filter matrix.

The dry ashing procedure was tested on each type of filter under study. It was felt that a minimum inclusion of organics in the carrier-free solution would lessen the possibility of interference in subsequent determinations and that the dry ashing procedure afforded a simple technique without danger of increasing the beryllium and uranium blanks.

In many instances this technique was not useful. Specifics are discussed below.

The investigations and finalized procedures are written such that they will prove satisfactory under the most disadvantageous analytical conditions that could be realistically anticipated. These anticipated disadvantageous conditions involve inclusion of one or more of the volatile radionuclides, and uranium or beryllium in the separation and determination scheme, and the possibility of a large amount of background dust being collected on the sampling media.

The following section of this report summarizes the various analytical investigations performed at Tracerlab, Incorporated and includes pertinent discussion and data from the two Quarterly Progress Reports issued previously.[1]

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[1] Quarterly Progress Report for June 15-August 31, 1960, Contract AF 29(601)-2852. Quarterly Progress Report for August 31-November 30, 1960, Contract AF 29(601)-2852.

C. EXPERIMENTAL RESULTS

1. Filter Dissolution Investigations

a. Pleated Filters

The pleated filters are TFA Type "S" filters, manufactured by the Staplex Company, Air Sampler Division. The filter is constructed from a pleated cellulose paper and is recommended for collection of particulate matter of ten microns or over in size.

Trial wet dissolutions of the pleated filters in nitric acid, hydrochloric acid or aqua regea failed to effect dissolution. Even treating a small amount of filter with large amounts of acids merely resulted in the swelling of the filter and did not appear to attack the structure of the filter. Further work on wet dissolution of this type filter was dropped in favor of investigating dry ashing techniques.

The conditions of dry ashing were dictated by the volatility of cesium, ruthenium and molybdenum. Cesium and molybdenum are known to be stable up to 450° C, but ruthenium remained to be investigated. Drying $RuCl_3$ carrier at 100° C indicated that the ruthenium compound present was $RuCl_3 \cdot 3H_2O$. Ashing this residue at 450° C for 48 hours resulted in a weight loss of 52 percent. However, further ashing produced no additional weight loss. The conversion of the $RuCl_3 \cdot 3H_2O$ to Ru_2O_3 stoichiometrically fits this situation and is assumed to be the case here. Further work using ruthenium metal instead of the trichloride resulted in no apparent weight loss.

With the optimum dry ashing temperature established, the dry ashing of the pleated filter proceeded. It was ashed at 425° - 450° C for 48 hours in a muffle furnace. No more shrinkage was observed after the first 24 hours. A perfect miniature replica 1.5 inches in diameter was produced. The ash was then given three fuming nitric treatments without observable reaction. (Perchloric acid was not used in experimentation since it would volatilize any Ru present and could not be used in actual samples.) The ash was then transferred to a teflon beaker and treated with HNO_3-HF mixtures. Solution was

effected with difficulty. The solution was transferred to a volumetric flask and diluted to the mark for use in further experimentation.

Subsequent dry ashing of the pleated filters did not produce consistent results. The shrinkage of the filter was noted in only one out of four cases. It is assumed that this effect is due to variations in the quantity of the adhesive binder present.

It was observed that at about 250° C the filters would proceed to ash at red heat for a few minutes until all the volatiles were expended. The red heat charring occurred even when the filter was placed in a cold oven and the temperature was raised by 50° C over a period of two hours.

Although the dry ash method reduced the weight of the filter from about eight grams to about four grams the auto-induced high temperature charring renders the procedure useless when any of the volatile elements cesium, ruthenium or molybdenum are to be analyzed.

An HF-HNO_3 dissolution of the pleated filter was then investigated and used successfully on four filters. Repeated HNO_3-HF digestions are used to completely dissolve the filter. The HF is evaporated and the solution diluted to volume in a 250 ml flask using 3 $\underline{N}$ HNO_3 and H_2O. The finalized procedure is appended.

b. Carbon Filters

The activated charcoal and carbon filters were found to be insoluble in the ordinary reagents employed on paper filters. Due to this fact and the large amount of carbon present (59 grams per charcoal filter, 2 grams per carbon trap) the dry ash procedure was used to destroy the carbonaceous material. The dry ashing was extremely slow at 450° C, so the temperature was increased to 500° C. Ashing was then completed in 18 hours.

Subsequent to the dry ashing the residue was transferred to a teflon beaker and treated with repeated portions of HNO_3-HF. A small amount of residue remained which eventually was leached several times and removed by centrifugation before transferring to a volumetric flask and diluting to volume.

The procedure discussed above gave rise to a variety of analytical problems. The dry ash often took as long as forty-eight hours and in no case gave a clean ash. A large amount of siliceous-like material usually remained semi-fused to the beaker in which the carbon had been ashed. This residue dissolved with difficulty and often had to be leached and discarded.

Air was injected over the sample during the ashing in an attempt to increase the combustion rate. However, as in the case of the pleated filter, auto-combustion took place at red heat and even an extremely slow stream of air tended to disturb the light ash present.

Due to the shortcomings of the dry ashing technique, an attempt was made to destroy the carbonaceous material with atomic oxygen. Point heating in the pyrex combustion tube caused the tube to break before any observable ashing had taken place. A vycor combustion tube was constructed and substituted for the pyrex tube. This type of combustion tube withstood the point heating effect without damage. However, the surface of the carbon particles quickly formed a protective ash which seemed to inhibit further reduction of the particles. After about one-half hour no further ashing of the filter was observed and the destruction of the filter was negligible.

An acid leach of the carbon was investigated next. Concentrated nitric and hydrochloric acids and 6 N HNO_3 and 6 N HCl were tried. The 6 N HCl appeared to be the best leach media. The concentrated acids leached a large amount of the siliceous material commented on earlier and caused difficulty in obtaining a clear solution. An HF treatment of the leachate is required to insure complete dissolution of the leached radionuclides.

The finalized procedure is appended.

c. Fiberglass Filters (MSA 1106B)

The 1106B filter is manufactured by the Mine Safety Appliance Company, Pittsburgh, Pa. It is constructed of glass microfibers and natural esparto glass and contains about 7 percent organic binder material. Filter disks 4" in diameter were used in the investigations discussed below.

Nitric and hydrochloric acids failed to affect this filter and dry ashing at 450° C resulted in no observable change in the filter structure. However, treatment of the filter in HNO_3-HF mixtures readily renders the filter soluble. Three treatments were found to be necessary for complete dissolution. The first few attempts to dilute the resulting solution to 100 mls for sample aliquotting resulted in obtaining a cloudy solution. Subsequent experimentation varied the dissolution technique; it was found that including several aqua regia treatments after the initial HNO_3-HF treatments yielded a clear, colorless solution upon subsequent dilution to volume.

Chemical analysis of these filters indicated than an average of 16 mg of inert barium is present in the filter matrix necessitating correction of the chemical yield.

The procedure for the fiberglass filter dissolution is appended.

d. Millipore Filters

The Millipore filter is a thin cellulosic porous membrane material available in ten porosity grades. Type RA, pore size 1.2 microns, 47 mm diameter was used in these investigations.

Several dissolution methods appear to be suitable for the destruction of this filter. The dry ashing causes rapid ignition of the paper with the attendant possibility of partial or total loss of the sample. Wrapping the Millipore filter in a less explosive ashless paper such as Whatman No. 40 prevents this occurrence and gives a clean ash.

The rapid ignition of the paper can also be prevented by treating the paper with nitric acid prior to the ashing and is the method included in the appendix.

Although the Millipore filter is readily soluble in nitric acid, the dry ashing procedure has been included to destroy the volatile carbonaceous material which could be a potential interference in the subsequent beryllium or uranium determination.

An $HF-HNO_3$ treatment has also been included in the finalized procedure to insure complete dissolution of any siliceous material present.

Of the various filters investigated for air effluent collection, the Millipore filter is the easiest to get into solution and offers the most consistent fiber matrix.

e. Sticky Paper

Sticky paper is a cellulosic paper with one gummed surface. The papers used in the following experiments were 13" by 13".

Treatment of this paper in ordinary dissolution reagents such as hydrochloric or nitric acid effects dissolution of the paper base but not the adhesive which, under actual sampling conditions, is likely to contain the majority of the particles of interest.

The dry ashing procedure is effectively used on this paper. To prevent rapid combustion of the filter, the ashing temperature should be raised to 450° C at 50° C intervals starting at 200° C. Complete ashing can be accomplished in ~6 hours. The oven temperature is increased only after smoking has stopped at any given setting. If this process is not followed, flaming or spattering of the sample will occur.

The dry ashing procedure leaves a light grey residue which is unaffected by HNO_3 or aqua regia treatments. The ash is transferred to teflon and treated with 10 mls HF and 2 mls HNO_3 to effect dissolution. This step requires an additional hour. Multiple samples may be run by this method.

This method was successfully carried out on six sticky papers and is included in the appendix.

f. IPC Filters

The IPC filters are composed of cotton linter fibers attached to a cotton skrim backing and impregnated with Kronisol, a di-butoxyethyl phthalate

organic compound which improves the retention properties of the filters. The paper has a density of about 14 grams per square foot and retains essentially 100 percent of spherical particles 0.02 microns or greater in diameter.

Although this type of paper was not originally specified for investigation by AFSWC, its widespread use as a sample collector in work of this type and the variety and extent of experience which Tracerlab personnel have had in dealing with it, later dictated that it be included as a possible effluent sample collection medium. Discouragingly large beryllium and uranium blanks on some of the other filter paper types also led to the investigation of this paper.

Dry ashing four square feet of this paper overnight at 450° C. reduces the paper to a clean ash.

Subsequent $HF-HNO_3$ treatments insure complete dissolution and yield a clear, colorless solution upon dilution to a known volume.

The reagent blank is extremely low for this dissolution. Unfortunately the uranium blank in the paper is about 0.025 µg per square foot which restricts the resolution of small uranium samples from the blank uranium.

The procedure used at Tracerlab for the dissolution of IPC paper is appended.

2. Beryllium Analysis

a. Reagent Blank Investigations

Due to the low level of beryllium anticipated in the reagents, blank filters and exposed filters, the most sensitive available method for beryllium determination was used. This method employs a 3.4 meter Ebert spectrograph and is discussed in detail in the appended procedure. Beryllium content of the reagents tested are tabulated below.

TABLE 1. BERYLLIUM IN REAGENTS

Reagent	µg/Liter Be
Hydrofluoric Acid (B and A Reagent Grade)	0.025
Nitric Acid (B and A Reagent Grade - redistilled)	< 0.005
Nitric Acid (B and A Reagent Grade)	< 0.005

b. Unexposed Filter Paper Blank

Beryllium content of clean filters was determined by emission spectroscopy as per the appended procedure. Table two below summarizes these results.

TABLE 2. BERYLLIUM CONTENT OF UNEXPOSED FILTERS

Filter Type	A	B	C	D
	µg/ml x 10^3	µg/ml x 10^3	µg/Filter x10^3	µg/Filter x10^3
Charcoal	0.75	0.92	75.	92.
Staplex(Pleated) No. 2	9.0	9.8	2250.	2450.
Staplex(Pleated) No. 3	14.1	13.7	3525.	3425.
1106B No. 1	0.1 (est)	0.1 (est)	<10.	<10.
1106B No. 2	0.1 (est)	0.1 (est)	<10.	<10.
Millipore No. 4	0.88	0.94	4.4	4.7
Millipore No. 5	<0.1	<0.1	<0.5	<0.5
Sticky Paper No. 3	0.09	0.11	0.90	1.1
Sticky Paper No. 4	0.11	0.15	1.1	1.5

Columns A and B represent the amount of beryllium per milliliter of carrier-free solution. Columns C and D convert columns A and B to the amount of beryllium in the total solution.

c. Exposed Filter Blank

The beryllium blank from filters exposed in a Staplex sampler for approximately five hours at the Nevada Test Site was determined from a carrier-free dissolution. The filters were MSA 1106B fiberglass filters. Specially purified reagents were used with the exception of hydrofluoric acid which was standard Baker and Adamson reagent grade. Six filters were received and were labeled BDR (Blank Dust Run) Nos. 1 - 6. Uranium and radionuclide blanks were also ran from the carrier-free solution of these filters. Results of these analyses are tabulated later in this report.

Table three summarizes emission spectroscopic results for beryllium content of these filters. Five ml samples were aliquotted for these analyses.

TABLE 3 BERYLLIUM CONTENT OF EXPOSED FILTERS

SAMPLE NO.	Run 1 $\mu g/ml \times 10^3$	Run 2 $\mu g/ml \times 10^3$	Run 1 $\mu g/Filter \times 10^3$	Run 2 $\mu g/Filter \times 10^3$	Average $\mu g/Filter \times 10^3$
BDR-1A	0.7	0.6	70.	64.	67.
BDR-2A	1.0	1.1	98.	108.	103.
BDR-3A	0.7	0.9	74.	86.	80.
BDR-4A	0.8	0.7	80.	68.	74.
BDR-5A	0.7	0.5	68.	54.	61.
BDR-6A	0.5	0.6	52.	56.	54.

3. Uranium Analysis

a. Reagent Blank Investigations

The uranium content of the reagents used in the dissolution of filters and the uranium analysis was determined by mass spectrometry. The method used is discussed later in this report and appended.

The results of these analyses are as follows, presented in Table Four below.

TABLE 4 URANIUM IN REAGENTS

Commercial Reagent	Uranium Content
f HNO_3	10 Sm*/liter
HF	14 " "
H_2O (distilled)	4 " "
$HClO_4$	7 " "

Distilled Reagent	Uranium Content
f HNO_3	5 Sm/liter
$HClO_4$	2.5 " "

* Sm = Smidgeon = 10^{-9} g

The fluorometric determination of gross uranium was also investigated. This method for uranium determination is limited to samples of greater than 10^{-8} grams. This method is not as accurate as the mass spectrometric determination but for 10^{-8} grams or greater it complies with the 10 percent requirements.

The fluorometric method involves taking a suitable aliquot of the sample solution, drying in a platinum dish, fusing with NaF-LiF flux and determining the uranium fluorometrically against a standard uranium solution. Work done on this procedure included flux pellet blank studies and the determination of optimum fluxing temperatures, time, and cooling cycles. A Jarrell Ash Model 2600 fluorometer was used in these experiments.

Determination of uranium in an unknown solution provided by AFSWC Headquarters gave values of 4.6 μg/liter by fluorometry and 4.5 μg/liter by mass spectrometric determination.

It is anticipated that the fluorometric procedure will not lend itself directly to the determination of uranium due to the quenching from the large amounts of ash obtained from filters containing a high percentage of fiberglass (pleated and 1106B). However, in the case of the Millipore filter and sticky paper this procedure may be readily usable.

The procedure is appended for reference.

b. Unexposed Filter Paper Blank

The uranium content of clean filters was determined by mass spectrometry according to the appended procedure. Table Five below tabulates the results of these investigations. The aliquot from the carrier-free solution used for this analysis represented .05 of the total solution in each case.

TABLE 5. URANIUM CONTENT OF UNEXPOSED FILTERS

SAMPLE	Atom Percent Values		μg U per Total Filter x 10^3		
	U^{235}	U^{238}	U^{235}	U^{238}	Total U
Staplex No. 2	0.80 ± 0.01	99.20 ± 0.01	28.2 ± 0.97%	3690. ± 1.52 %	3720. ± 1.50 %
Staplex No. 3	0.75 ± 0.02	99.25 ± 0.02	23.9 ± 2.94%	3160. ± 1.25%	3185. ± 1.25%
Millipore No. 1	2.54 ± 0.20[a]	97.46 ± 0.20	0.0197 ± 9.0%	0.754 ± 4.5 %	0.774 ± 4.39%
Millipore No. 2	0.72[b]	99.28[b]	0.0078[b]	1.08 ± 2.21%	1.09 ± 2.21%
Sticky Paper No. 5	0.79 ± 0.02	99.21 ± 0.02	0.0379 ± 2.72%	4.76 ± 1.26%	4.80 ± 1.25%
Sticky Paper No. 6	1.06 ± 0.01	98.94 ± 0.01	0.0651 ± 2.02%	6.16 ± 2.30%	6.23 ± 2.25%
Act. Charcoal No.1	25.28 ± 0.25	74.32 ± 0.25	394. ± 1.73%	1173. ± 2.18%	1573. ± 1.68%
1106B No. 1	0.93 ± 0.05	99.07 ± 0.05	3.98 ± 4.34%	430. ± 7.2 %	434. ± 7.1 %
1106B No. 2	0.86 ± 0.01	99.14 ± 0.01	3.38 ± 1.06%	396. ± 1.30%	399. ± 1.29%

a) High 235 background. Percent assumed to be high.

b) High 235 background. Percent assumed to be correct.

c. Exposed Filter Paper Blank

The uranium blank in filters exposed on the Nevada Test Site was determined by mass spectrometry of five ml aliquots from the sample solution. The history of the filters was discussed previously under paragraph C.2.c. Table Six tabulates the results of these analyses.

TABLE 6 URANIUM CONTENT OF EXPOSED FILTERS

SAMPLE NO.	Atom Percent U^{235}	Atom Percent U^{238}	μg/ml x 10^3 Total U	μg/Filter x 10^3 Total U
BDR-1	0.74 ± 0.01	99.25 ± 0.01	4.24 ± 2.0 %	424. ± 2.0 %
BDR-2	0.72 ± 0.02	99.28 ± 0.02	4.26 ± 3.34%	426. ± 3.34%
BDR-3	1.09 ± 0.05	98.91 ± 0.05	4.10 ± 6.05%	410. ± 6.05%
BDR-4	0.87 ± 0.02	99.13 ± 0.02	4.10 ± 4.27%	410. ± 4.27%
BDR-5	0.78 ± 0.01	99.22 ± 0.01	4.62 ± 3.3 %	462. ± 3.3 %
BDR-6	0.79 ± 0.01	99.21 ± 0.01	4.46 ± 3.79%	446. ± 3.79%

4. Radionuclide Analysis

a. Separation Procedure

In accordance with the work statement, a separation scheme capable of separating U, Be, Ba, Sr, Mo, Ru, Cd, Cs, Y and Np in good yield from a single filter was designed and investigated.

Carrier-free aliquots for uranium and beryllium were employed to insure a minimum contribution of these elements from reagents. In addition, redistilled reagents were used in the dissolution procedures.

The first separation scheme investigated is abstracted as follows: Precipitate the barium and strontium as nitrates, carry neptunium on yttrium fluoride, distill over the ruthenium in perchloric acid and separate cesium, cadmium, and molybdenum on a Dowex 1-X10 anion resin column. This procedure was tried and some difficulty encountered. The yield on visual observation appeared satisfactory.

On subsequent trials, this scheme continued to present problems so the scheme was altered as follows: Precipitate the barium and strontium as sulphates and distill the ruthenium as the tetraoxide from perchloric acid. Evaporate the residue in the flask to wet dryness and dissolve the carriers in 12 N HCl. Load the carriers onto a Dowex 1-X10 anion exchange column and elute the Cs and Y with continued additions of 12 N HCl. Add 1 N HCl-0.1 N HF to elute the Np, add 3 N NH_4OH to elute the cadmium and add NH_4OH-$NH_4C_2H_3O_2$ to elute the molybdenum.

This procedure was used in the analysis of the BDR (Blank Dust Run) samples obtained from the Nevada Test Site. The procedure is detailed in the appendix.

The BDR samples described in paragraph C.2.c. were analysed for the long-lived fallout isotopes, strontium-90 and Cesium-137. Results of these analyses are tabulated below.

TABLE 7 BACKGROUND ANALYSIS FOR CS-137 AND SR-90

SAMPLE	DPM/Filter	
	Sr-90	Cs-137
BDR-1*	4.90 ± 4.4%	<7.2
BDR-2*	<1.4	<5.3
BDR-3	1.59 ± 9.7%	<1.2
BDR-4	<1.5	<1.2
BDR-5	<1.4	2.03 ± 9.0%
BDR-6	1.49 ± 10.1%	<1.0

* Low Yield

The quoted DPM values were obtained by correcting the absolute counting rate for counter efficiency, counter background, chemical yield, precipitate thickness, and fraction of the filter used in the analysis. No blank corrections were made. One-half of the filter was used in each case, the other half being used for beryllium and uranium analysis. The samples were counted on Tracerlab CE-14 low background counters. The chemical yields were in the range of 65 - 75 percent with the exceptions noted in the table, which were about 10 percent yield. The major portion of these samples was lost due to a pressure "back-flash" in the ruthenium distillation flask.

The samples were counted on days 334 - 336, 1960, and were not corrected for decay.

Other isotopes decontaminated and counted were Cd^{115}, Ba^{140}, Mo^{99}, Ru^{106}, and Y^{91}. In each case a limited value was obtained indicating a negligible background of these isotopes. These investigations also served to check the radiochemical purity of the carriers and reagents used in the chemical procedures. Table 8 below tabulates the results of these investigations.

TABLE 8. RADIONUCLIDE BACKGROUND ANALYSIS

SAMPLE	DPM/Filter				
	Cd-115	Ba-140	Y-91	Mo-99	Ru-106
BDR-1*	< 6.0	< 1.0	< 3.8	< 6.3	< 7.0
BDR-2*	< 8.0	< 1.8	< 7.0	< 6.0	N. D.
BDR-3	< 1.3	< 1.9	< 1.1	< 1.6	N. D.
BDR-4	< 3.0	< 1.3	< 1.1	< 1.5	< 2.6
BDR-5	< 2.0	< 1.2	< 1.4	< 1.4	< 2.3
BDR-6	< 1.5	< 1.1	< 1.1	< 1.3	< 1.2

* Low Yield

N. D. - Not Determined

The quoted DPM values were calculated in the same manner as those in Table 7.

It appears that no background corrections are necessary when effluent sampling conditions similar to those of the BDR sample collection are present.

b. Decontamination Procedures

The decontamination procedures used were those already in use at Tracerlab. They have been developed over a period of years for use in fallout studies, reactor effluent studies, activation analysis, isotope applications and tracer studies. These procedures are considered adequate to give pure products in fresh or old nuclear debris.

The procedures are appended as used in the BDR analysis.

c. Counting and Calculation

The counting and calculation procedures vary with the type of analysis used, the origin of the sample and the purpose for which the data is to be used. The appended procedures are considered applicable to analysis of air effluent from the Tory reactor but may be modified as required by the Air Force Special Weapons Center.

5. Mass Spectrometric Analysis for Isotopic Uranium

For samples below 10^{-8} grams, the uranium aliquot from a carrier-free sample solution is spiked with U^{233} tracer and the uranium analyzed mass spectrometrically. The yield of the uranium is then determined from the U^{233} peak and the sum of the other uranium peaks corrected accordingly. If U^{233} is to be determined in the sample, the sample is split equally into two portions. One portion is spiked with U^{233} and the uranium determined as outlined above. The other fraction is run spike-free for the determination of U^{233} and the chemical yield

determined through the analysis of relative peak heights. This procedure is capable of detecting uranium on the 10^{-9} gram level within ± 10 percent.

6. Particle Investigation

Since most of the Tracerlab particle isolation and investigation methods depend on autoradiography and the sample filters were activity free, no specific investigations were performed to isolate particles. However, a literature search was performed and the procedures for particle isolation and investigation were studied for adaptation to analyzing the air effluent of the Tory reactor.

Procedures for particle detection, dispersion, isolation, decontamination, and determination of physical and radiochemical properties are appended.

Minor revisions may be necessary if the particle collection media has properties widely divergent from those previously encountered in work of this nature at Tracerlab.

It is anticipated that Institute of Paper chemistry paper, MSA 1106B filters or Millipore filters will readily lend themselves to analysis of collected particulate debris.

D. CONCLUSION

The foregoing discussion and appended procedures satisfy all requirements of Contract AF 29 (601)-2852.

1. Methods of dissolution of pleated filters, carbon traps, fiberglass filters, Millipore filters, sticky paper and IPC filters have been investigated and a dissolution procedure developed for each of the above filters.

2. A rapid separation scheme capable of separating U, Be, Ba, Sr, Mo, Cd, Y, Ru, Cs, and Np in good yield has been successfully developed and tested.

3. Existing decontamination procedures were applied to each of the above elements and found satisfactory.

4. The detection and determination of beryllium and uranium on the 10^{-9} gram level within ten percent error limits has been accomplished.

5. The background level of beryllium, uranium and radionuclides present in filter papers, reagents and the environment were determined.

6. The isotopic composition of uranium was determined by mass spectrometry and the feasibility of studying particle properties as a reactor diagnostic tool investigated.

7. The problems involved in air sampling with the various filter types are now more fully understood and the practical problems involved in the subsequent radiochemical analysis have been enumerated.

The following recommendations are derived from an examination of the information and data contained in this report.

E. RECOMMENDATIONS

The unexposed filter blank studies indicate that the 1106B, and Millipore filters and sticky paper have the lowest beryllium blank and the Millipore filter and sticky paper have the lowest uranium content. The high beryllium blank of the charcoal and Staplex pleated filters and the high uranium blank of the Staplex, charcoal and 1106B filters indicates that they should not be used as a sampling medium when analyzing for low levels of these elements. The Millipore filter and sticky paper remain as the optimum sampling medium insofar as beryllium and uranium background are concerned.

The chemical difficulties involved in dealing with the charcoal and Staplex filters and the inclusion of inert barium in the matrix of the 1106B filter also indicate that these filters should not be used for sampling beryllium and uranium.

The beryllium and uranium content of the reagents used in the filter dissolution is not a problem. However, the air effluent samples will have to be corrected for the atmospheric background of beryllium and uranium. There is no significant atmospheric background of radionuclides and these should not require correction.

In short, it is recommended that Millipore filters and sticky paper be used as sampling media, a dry-ash and $HF-HNO_3$ dissolution using highly purified reagents be performed and the sample diluted to a known volume. The beryllium and uranium should be analyzed emission spectrometrically and mass spectrometrically respectively from aliquots withdrawn from the carrier-free solution. The radionuclides should be analyzed according to the appended separation, decontamination and counting procedures.

F. APPENDIX

1. Filter Dissolution Procedures

a. Staplex (Pleated) Filter Dissolution

1. Place the filter in a 600 ml teflon beaker. Add 25 ml HF, 10 ml HNO_3 and evaporate to wet dryness.
2. Repeat step 1 three times.
3. Add 2 ml saturated H_3BO_3 and 10 ml H_2O. Evaporate to wet dryness.
4. Add 10 ml HNO_3 and 5 ml HCl. Evaporate to dryness.
5. Repeat step 4 twice.
6. Add 25 ml HNO_3 and 5 ml H_2O to the beaker, bring to a boil and transfer to a 250 ml volumetric flask (note a). Complete the transfer with 5 - 10 ml portions of 3 N HNO_3.
7. Add 100 ml 3 N HNO_3. Warm the flask gently until a clear solution is obtained. After cooling to 20° C, make up to the mark with H_2O.

Note (a) If a smaller flask is used or if the acid concentration is >4 N, the sample tends to salt out.

Total time for 2 samples - 5 hours.

Total reagents in dissolution of each filter: HNO_3 - 70 mls, HCl - 15 mls, H_2O - 10 mls, HF - 100 mls, H_3BO_3 - 2 mls.

b. Carbon Filters

1. Remove the carbon from its container and place in a 600 ml Pyrex beaker. Add 300 mls 6 N HCl and heat to 50° C. Hold at this heat for 30 minutes.
2. Decant the major portion of acid into another 600 ml beaker. Add another portion of 6 N HCl to the carbon and again heat and decant the acid.

3. Add 2 N HCl to the carbon and filter the acid through a Whatman No. 40 paper. Transfer the carbon to the filter and wash thoroughly with 2 N HCl.

4. Evaporate the accumulated acid containing the leached activity to about 100 mls and transfer to a 250 ml teflon beaker. Add 20 mls HF, 50 mls HNO_3 and evaporate to a small volume.

5. Repeat the HNO-HF treatments until a clear solution is obtained. (Two treatments should suffice.)

6. Evaporate the solution until a moist residue remains. Add three ml saturated H_3BO_3 and ten ml 3 N HNO_3.

7. Transfer the sample to a 250 ml volumetric flask using successive washes of 3 N HNO_3.

8. Fill the flask to approximately 200 mls using 3 N HNO_3. If the solution is not clear, warm the flask gently in a hot water bath until a clear solution is obtained.

9. Cool the sample to 20° C and dilute to the mark with 3 N HNO_3.

Total time for two samples - 8 hours.

Total reagents in dissolution of each filter:
HCl - 350 ml, HNO_3 - 200 mls,
H_2O - 500 ml, HF - 60 mls, H_3BO_3 - 3 mls.

c. Fiberglass Filter (1106B) Dissolution.

1. Place the fiberglass filter in a 50 ml teflon beaker. Wet the filter with 4 ml. HNO_3 (note a) add 10 mls HF and evaporate the sample to wet-dryness.

2. Add 2 ml HNO_3 and 10 ml HF and evaporate to dryness.

3. Repeat step 2 twice.

4. Add 2 ml saturated H_3BO_3, 5 ml H_2O and evaporate to dryness.

5. Add 4 ml HNO_3, 2 ml HCl and evaporate to dryness.

6. Repeat step 5 twice.

7. Rinse down the sides of the beaker with 3 ml HNO_3 and evaporate to dryness.

8. Take up the sample in 10 ml HNO_3 and transfer to a 100 ml volumetric flask. Complete the transfer using 5 - 2 ml portions of 6 N HNO_3.

9. Add approximately 60 ml 6 N HNO_3 and gently warm the flask until a clear solution is obtained.

10. Cool the sample to 20° C and make up to the mark with H_2O.

Note (a) Unless the entire filter has been wetted with HNO_3, a violent reaction will occur with the addition of HF.

Total time for two samples - 3 hours.

Total reagents per filter: HNO_3 - 25 mls, HF - 40 mls, HCl - 6 mls, H_2O - 5 mls, H_3BO_3 - 2 mls.

d. Millipore Filter Dissolution

1. Place the filter in a 50 ml teflon beaker (note a) and add 4 ml HNO_3. Place on a hot plate set at low heat and evaporate to wet-dryness. Add 2 ml HNO_3 and again take the sample to wet-dryness.

2. Add 2 ml HF and 1 ml HNO_3. Evaporate just to dryness and repeat the treatment with 2 ml HF - 1 ml HNO_3.

3. Add 2 drops saturated H_3BO_3, 2 ml H_2O and evaporate to dryness.

4. Take the sample up in 2 ml HNO_3, bring to a boil and transfer to a 50 ml quartz beaker using 3 - 1 ml portions of HNO_3 to complete the transfer. Evaporate the sample to dryness. Place the quartz beaker in a 450° C oven for one hour to complete the destruction of organic material.

5. After allowing the beaker to cool, add 2 ml HNO_3 and 1 ml HCl. Take the sample to dryness and repeat the aqua regia treatment once.

6. Take the sample up in 2 ml HNO_3. Bring to a boil and transfer to a 10 ml volumetric flask using 3 - 2 ml portions of 6 N HNO_3 to complete the transfer. After cooling the solution to 20° C, make up to the mark with H_2O.

Note (a) These filters have a static charge on them and are difficult to handle before the first addition of HNO_3.

Total time for 2 samples - 3 hours.

Total reagents per filter: HNO_3 - 17 mls, HF - 4 mls, HCl - 2 mls, H_2O - 1 ml.

e. Sticky Paper Dissolution

1. Put the sticky paper in a 100 ml Pyrex beaker and place in a cold muffle oven. Set the thermostat at 250° C. After the oven has reached 250° C continue heating at this temperature until the sample has stopped smoking.

2. Raise the oven temperature to 450° C at 50° intervals after smoking has stopped at any given setting. Leave the sample in the oven for 1 hour after 450° C has been reached.

3. Add 2 ml HNO_3 to the sample, bring to a boil and transfer to a 50 ml teflon beaker using 2 - 2 ml portions of HNO_3 to complete the transfer.

4. Add 5 ml HF and evaporate the sample to wet-dryness.

5. Repeat step 4 once.

6. Add 2 ml HNO_3, 1 ml HCl, and 0.5 ml H_3BO_3, and evaporate the sample to wet-dryness.

7. Add 2 ml HNO_3 and evaporate to wet-dryness.

8. Add 2 ml HNO_3, bring the sample to a boil and transfer to a 50 ml volumetric flask. Complete the transfer with 5-2 ml portions of 6 N HNO_3. Add 35 mls 6 N HNO_3 and warm gently until a clear solution is obtained. Cool the sample to 20° C and make up the mark with H_2O.

Total time for 2 samples - 10 hours.

Total reagents per sample: HNO_3 - 10 mls, HCl - 1 ml, HF - 10 mls, H_3BO_3 - 0.5 ml.

f. IPC Filter Dissolution

1. Place the filter in a 400 ml beaker, cover the beaker with aluminum foil. Poke six pencil sized holes in the foil and place the beaker in a 450° C oven. Ash overnight.
2. Transfer the ash to a 250 ml teflon beaker using HNO_3 as a wash.
3. Add 3 mls HNO_3 and 1 ml HF. Evaporate to wet-dryness and repeat HNO_3-HF treatments until complete dissolution is effected.
4. Evaporate residue to wet dryness, add 0.5 ml H_3BO_3, 20 ml of 4 N HNO_3 and heat. Transfer to a 100 ml volumetric flask using 4 N HNO_3 washes.
5. Cool the transferred sample to 20° C and dilute to the mark with 4 N HNO_3.

Time (~~e~~xcluding dry ash) - 3 hours.

Reagents used: HNO_3 - 15 ml, 4 N HNO_3 - 100 mls, HF - 5 mls.

2. Beryllium Analysis

a. Fluorometric

1. Pipette a suitable aliquot of the carrier-free sample solution into a 100 ml beaker. Add 2 mg Al carrier, 30 ml of digestion mixture (80 percent HNO_3 - 20 percent $HClO_4$) and evaporate the samples to ~2 ml.

2. Transfer the sample to a 40 ml centrifuge cone and add 1 ml of mercaptoacetic acid. Bring to pH 8 using NH_4OH and centrifuge the resulting precipitate. Discard the supernate and wash the precipitate with a solution prepared as follows: Bring 2 ml mercoptoacetic acid to a pH of 8 using conc. NH_4OH and dilute to 100 mls using solution "A". Prepare solution "A" by bringing 10 ml conc. HCl to a pH of 8 with conc. NH_4OH and diluting to 1000 ml with H_2O. The wash solution is stable one day.

3. Continue washing and centrifuging the precipitate until the supernate is clear and the precipitate is white.

4. Dissolve the precipitate in 5 ml HCl and transfer to a 25 ml volumetric flask using 1 percent $HClO_4$ as a wash solution.

5. Add 1.5 ml NaOH-DTPA-TEA (note a) and one drop quinine sulphate indicator (note b). Titrate the sample to the fluorescent end-point with 72 percent $HClO_4$, using ultra-violet light.

6. Defluoresce the sample using 1 N NaOh. Add 5 ml piperidine buffer (note c) and 1 ml morin (0.015 percent w/v in 40 percent ethanol).

7. Dilute to the mark with H_2O, mix thoroughly and allow the sample to react for 5 minutes.

8. Allow the sample to come to a constant temperature and read on a Turner Associates Model 110 fluorometer using 2A + 47B primary filters and 2A-12 + 58 secondary filters.

Note (a) - NaOH-DTPA-TEA solution is prepared as follows: Dissolve 165 grams of sodium hydroxide in 400 ml of water. Dissolve 2 ml of colorless triethanolamine (TEA) and 5 grams of purified DTPA in the alkaline solution and dilute to 500 ml. Store in a polyethylene bottle.

Note (b) Quinine indicator is prepared as follows: Dissolve 50 mg of quinine sulfate in 500 ml of water containing 10 ml of 72 percent $HClO_4$.

Note (c) Piperidine buffer is prepared as follows: Transfer 15.0 grams of purified DTPA to a 500 ml volumetric flask with about 200 ml of water. Add 75.0 ml of redistilled piperidine, stopper the flask and swirl under a stream of cold water until cool. Add a solution of 20 grams of anhydrous sodium sulfite in 150 ml of water and dilute to 500 ml. Store in a tightly stoppered bottle with a polyethylene - lined screw cap.

b. Emission Spectrometric Beryllium Analysis

1. A suitable aliquot from the carrier-free solution of the sample filter is concentrated to one ml.
2. A 50λ drop is placed on each of two graphite electrodes and sparked.
3. Standards are prepared and ran in the same manner. The intensity of the spectral lines on a photographic plate are compared with the standard to determine the amount of beryllium in the sample
4. If the beryllium is not identified on the 2 - 50λ samples, the sample solution is concentrated further and re-ran.

Note - This method employs a 3.4 meter Ebert spectrograph with a prism replacing the diffraction grating. The sensitivity may vary from run to run depending on the level of contaminating elements present. If a large amount of contaminants are present a Be - Al co-precipitation may be necessary prior to loading the samples on the electrode. This decontamination method is detailed in the method for determining beryllium by fluorometry.

3. Uranium Analysis

a. Fluorometric Uranium Analysis

The fusion dishes are formed from satin finish 90 percent Pt - 10 percent Ir alloy disks (0.015" thick by 0.748 ± 0.001" dia.) in the special forming die (0.750" dia.). The new dishes are cleaned by boiling in a 1 to 1 mixture of sulfuric and nitric acids and then are rinsed thoroughly in water and distilled water. They are then fused twice and washed before their initial use.

The fluoride pellets from the previous run are discarded. The dishes are cleaned by:

1. Boiling once in about 0.1 N HCl.
2. Boiling twice in distilled water.
3. Fusing the dishes with the NaF-LiF flux.
4. Repeating steps 1 and 2
5. Rinse each dish under the distilled water tap, using clean forceps to handle the dishes.
6. Place dishes face down on a clean paper towel to dry.

Lay out 20 dishes on the Nichrome wire screen-ring holder in the pattern shown on the "Fluorimetric Uranium Assay Data Sheet" (Figure 7, Ref. 1). The dishes should be placed as flat as possible and spaced evenly without touching each other.

The modified Fletcher burner used for fusion is as described in Ref. 1, Figure 3. The regulation of propane and compressed air are through standard gas regulators attached to a propane tank and compressed air supply respectively. Adjustment of the flame is by regulation of the air supply to give a stable flame of bout 900° C. Readjustment of the air is usually needed to maintain the proper flame size and temperature. This is easily done after experience with the burner is gained.

Start up the burner and then place the dish holder in position and ignite the dishes to a bright red heat for 30 seconds. Allow to cool in place.

Pipette the 100λ sample aliquots onto the dishes according to the "Data Sheet". First pipette the three aliquots of five samples. (Positions 3, 4, 5; 6, 7, 8; 9, 10, 11; 14, 15, 16; and 17, 18, 19 respectively.)

Next the three aliquots of the standard uranium solution are pipetted using a separate 100λ pipette. (Positions 2, 13, and 20).

Finally, using the pipette from the standard uranium solution, 100λ of standard uranium solution is added to one of each of the five samples. (Positions 4, 7, 10, 15, and 18.)

Dishes number 1 and 12 are fused without added solution in order to determine the blank reading due to the flux and the conditions of fusion. The 100λ micropipettes are cleaned in a pipette cleaner using successive washes of detergent soap solution, distilled water chromic-sulfuric acid solution, distilled water and anhydrous alcohol. They are then coated with "Desicote" dryfilm preparation or a similar film.

The standard uranium solution used for spiking and standard measurement is 0.050 μg U_3O_8 per ml. This gives an aliquot of 5 smidgeons and enables all readings on the fluorometer to be made on the 0.001 scale. With the multiplier phototube operating at about 400 volts, typical readings are as follows:

Scale:	0.001	Instrument standard set on 80 div.
Blank:	10.8 Divisions	
Uranium Sample:	15.0 Divisions	
Standard Solution:	76.0 Divisions	
Spiked Uranium Sample:	68.0 Divisions	

When all aliquots have been added to the dishes, place an infrared heat lamp over the group and slowly evaporate all samples to dryness.

Place the sample-ring on a ringstand under a Fisher burner. Manipulate the flame carefully over each dish so as to ash each sample. This must be done very cautiously to prevent spattering of the samples and cross-contamination.

Replace the holder over the ignited Fletcher burner and ignite the dishes to a bright red heat for 30 seconds. Allow to cool.

The flux powder from which pellets are formed for fusion is composed of 2 percent LiF - 98 percent NaF mixed intimately. This is made up in one pound batches in order to insure uniformity in day to day use. A satisfactory batch of flux will give a reading of 1 smidgeon or less per 0.4 gram pellet. The purest NaF available gives 0.27 smidgeon. (Ref. 3)

The TLW pellet-maker is custom fabricated from pyrex glass. It is adjusted to deliver a 0.40 ± 0.01 gram pellet. The pellet-maker is gently pressed for about ten times into the flux powder container, and a spatula is used to flatten the bottom of the pellet.

Jarrell-Ash T-S-L fluorflux pellets may be used in lieu of the custom pellets but each batch must be checked to insure that the blank is low.

A flux pellet is delivered to each sample dish and the samples are immediately fused for 3 minutes. The burner flame is maintained at proper height and temperature during the fusion by adjustment of the air. The flux will melt within one minute if the flame is correctly adjusted, and the fusion is continued for three minutes after the last of the flux has melted. At the end of the fusion, turn off the air and gas simultaneously, and allow the pellets to cool for twenty minutes.

After the cooling period the pellets are removed from the fusion dishes and the fluorescence is read on a Jarrell-Ash Fluorimeter. Each button may be removed by holding the dish with forceps and tilting so that the button slides into the receptacle of the fluorimeter. If the button does not loosen from the dish, invert it on a piece of glazed paper and tap the bottom with the forceps. The forceps may be used to place and withdraw each button after the fluorescence reading is taken. Any loose particles are removed from the receptacle between each background reading by aspirating with a nozzle-rubber bulb.

The Jarrell-Ash fluorometer is operated and maintained according to the supplied manual. To prepare for a series of readings push the receptacle slide to the front stop. The instrument reference source is now under the ultraviolet light and the meter circuit is switched to the 0.01 scale. The voltage is adjusted so as to give a reading of 80 divisions. With this setting, all sample, standard and spike readings will be on the 0.001 scale. Adjust the reference, zero, and blank readings as specified and read each sample consecutively, rechecking the adjustment readings between each sample button reading. Record all necessary data on the "Assay Data Sheet"

Calculations

Average the blank readings(B) and the 5.0 smidgeon uranium standard (S). Calculate the U_3O_8 fluorescence equivalent of the instrument reference standard. A continuous record should be kept of this value together with the photomultiplier operating voltage.

Symbols (Reference 2)

A = Average reading of unspiked sample
S = μg of U_3O_8 in spike
D = Reading of spiked sample
B = Average reading of blanks
C = Reading of 1 μg U_3O_8
E = Average reading of U_3O_8 spike solution
F = Instrument reference standard reading
0.85 = Conversion factor for U_3O_8 to U.
10^4 = Aliquot factor for 100λ from one liter.

Instrument Reference Standard (μg U_3O_8) = $\frac{FS}{E-B}$

Blank (mμg or smidgeon U_3O_8) = $\frac{1000\ BS}{E-B}$

μg U_3O_8 in 100λ aliquot of sample = $\frac{AS}{D-A} - \frac{B}{C}$

μg Uranium per liter of sample = $(0.85 \times 10^4)\ \frac{AS}{D-A} - \frac{B}{C}$

FLUOROMETRIC URANIUM ASSAY
DATA SHEET

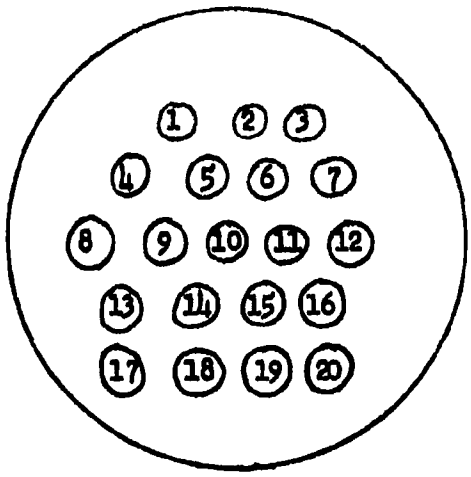
DATE_____ RUN NO._____

FUSION TIME_____

COOLING TIME_____

INST. STD. SET ON_____

FLUOROMETER VOLTAGE_____



(Readings below are on .001 scale unless otherwise noted).

Sample No., Position and Description		Reading		
1			1	Blank
2			2	100λ of Spike (Standard Uranium Solution).
3			3	100λ sample No. 1
4			4	100λ sample No. 1 plus 100λ Spike
5			5	100λ sample No. 1
6			6	100λ sample No. 2
7			7	100λ sample No. 2 plus 100λ Spike
8			8	100λ sample No. 2
9			9	100λ sample No. 3
10			10	100λ sample No. 3 plus 100λ Spike
11			11	100λ sample No. 3
12			12	Blank
13			13	100λ Spike
14			14	100λ sample No. 4
15			15	100λ sample No. 4 plus 100λ Spike
16			16	100λ sample No. 4
17			17	100λ sample No. 5
18			18	100λ sample No. 5 plus 100λ Spike
19			19	100λ sample No. 5
20			20	100λ Spike

REFERENCES ON FLUOROMETRIC URANIUM ANALYSIS

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4. Fluorometric Determination of Uranium by S. Kahn, Tracerlab, Inc., Western Division.

5. Maximum Permissible Internal Dose of Radionuclides: Recent Changes in Values. K. F. Morgan. Nuclear Science and Engineering, Vol. 1, No. 6, December, 1956.

6. National Bureau of Standards, Handbook 52.

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b. Mass Spectrometric Uranium Analysis

1. Pipette a suitable aliquot from the carrier-free sample solution into a quartz beaker containing ~4 x 10^{-9} grams of U-233 spike.

2. Heat the sample to dryness, allow to cool and then transfer the sample to a 12 ml centrifuge cone with 2 ml of acid deficient saturated ammonium nitrate reagent (note a).

3. The uranium is separated by extracting four times at five minutes each with 2 ml volumes of pre-extracted di-ethyl ether (note b).

4. Scrub each organic phase twice with saturated ammonium nitrate reagent and the combined ether phases are collected in another cone.

5. The uranium is extracted from the combined ether phases three times at 10 minutes each with 2 ml volumes of redistilled water.

6. Collect the water fractions in a 10 ml beaker containing 1 drop HNO_3 and 3 drops HCl.

7. Heat the beaker to moist dryness and treat the residue with aqua regia until it is completely destroyed. Add one drop of HNO_3 to the beaker and evaporate to dryness.

8. Transfer the uranium to a quartz cone with 2 N HNO_3 and heat to dryness under a 250 watt infrared heat lamp.

9. Wash the uranium from the cone and mount on a filament using ~5λ x 0.5 N HNO_3 for the transfer.

Note (a) The saturated ammonium nitrate is prepared by dissolving commercial analytical grade ammonium nitrate crystals in warm redistilled water and acidifying to 2 N HNO_3 with distilled concentrated nitric acid.

Note (b) The di-ethyl ether is pre-extracted with an equal volume of redistilled water for 10 minutes.

4. Radionuclide Analysis

a. Separation Procedure for Ba^{140}, $Sr^{89, 90}$, $Ru^{103, 106}$, $Cs^{136, 137}$, Y^{91}, Mo^{99}, Cd^{115} and Np^{239}.

1. Pipette an accurately known amount of Ba, Sr, Cs, Cd, Mo, Ru and Y carriers into a 250 ml beaker. (Approximately 20 mg of each carrier except Y should be used. Use ~10 mg of Y carrier.) Add ~500 DPM of standardized Np^{237} tracer.

2. Add the activity solution from a carrier-free dissolution of the sample filter, add 5 ml HNO_3, 3 ml H_2SO_4 and evaporate to wet dryness on a hot plate set at low heat. Use a speedy-vap cover during the evaporation.

3. Transfer the sample to a 40 ml centrifuge cone using 4 N HNO_3 washes. Centrifuge for 3 minutes and decant the supernate into a ruthenium distillation flask.

4. Wash the $BaSO_4$ and $SrSO_4$ with 4 mls of 4 N HNO_3, centrifuge and decant the supernate into the distillation flask. Save the sulphates for step 15.

5. Add 20 ml HNO_3 to the flask. With the still head in place, boil until only a few milliliters of solution remain in the flask. Add three ml of HCl.

6. Prepare a 40 ml centrifuge cone containing 10 ml of freshly prepared 12 N NaOH and place in an ice bath. A system to provide air flow through the distillation apparatus must be available.

7. To the sample in the distillation flask add 10 mg I^- carrier, 3 ml HNO_3 and 4 ml $HClO_4$. Connect the still head and start air flowing through the apparatus at about 2 bubbles per second. (See the ruthenium decontamination procedure for a more detailed study of the distillation.)

8. Heat the solution in the flask gently with a micro burner flame to begin the distillation, collecting the distillate in the 12N NaOH. Continue the distillation until the dense

white fumes of $HClO_4$ have passed over for one minute after all the ruthenium tetraoxide has been distilled. Save the distillate and continue with step 8 of the Ru decontamination procedure.

9. Allow the distillation flask to cool and transfer the contents to a 40 ml centrifuge cone using 6 N HCl washes to effect the transfer. Add 2 drops bromine water and stir well. Place the cone in an ice bath and saturate the liquid with hydrogen chloride gas.

10. Allow the solution to come to room temperature and transfer to a prepared Dowex 1-X8 100 - 200 mesh anion exchange column 12 mm in diameter by 3 cm long. Collect the effluent containing the Cs and Y and continue adding HCl to completely elute these elements (15 ml should suffice). Save this fraction for step 14.

11. Elute the neptunium with 15 ml of 1 N HCl-0.1 N HF added in 2 ml increments. Wash the column with 5 ml 1 N HCl and collect this wash with the Np fraction. Continue with step a of the Np decontamination procedure.

12. Elute the Cd with 20 ml of 3 N NH_4OH added in 2 ml increments. Evaporate to about 10 mls and continue with step a, sentence two of the Cd decontamination procedure.

13. Elute the Mo by adding 20 mls of 6 N $NH_4C_2H_3O_2$ containing 5 ml of NH_4OH added in 2 ml increments and continue with sentence two, step c of the Mo decontamination procedure.

14. Make the solution from step 10 basic using 6 N NaOH. Centrifuge the $Y(OH)_3$ and decant the supernate containing the Cs into a clean 40 ml centrifuge tube. Wash the $Y(OH)_3$ with 5 mls of 0.1 N NaOH, centrifuge and add the wash to the Cs sample. Dissolve the $Y(OH)_3$ in 15 ml of 2 N HNO_3 and continue with step 18. Add enough HCl to the Cs solution to make it about 6 N in HCl and continue with step b of the Cs decontamination procedure.

15. Add 10 ml of fresh 1 M Na_2CO_3 to the barium and strontium sulphates and boil until the volume is reduced to five ml. Centrifuge and discard the supernate.

16. Add 1 ml of 6 N HNO_3 to the residue, add 3 ml of H_2O and slurry well. Centrifuge and decant the dissolved carbonates into a clean centrifuge cone. Retreat the residue as in step 15 above until the sulphates are completely converted to carbonates combining all the dissolved carbonates in one centrifuge cone.

17. Add 25 - 30 ml of yellow fuming HNO_3 to the combined carbonates and cool in an ice bath for 10 minutes. Centrifuge, decant, and discard the supernate. Continue with step b of the barium decontamination procedure.

18. Add 10 mg Zr carrier, 10 mg Sr carrier and 10 mg Eu carrier to the Y sample. Transfer to a 50 ml lusteroid centrifuge cone using water washes. Add 2 ml HF, stir well and let stand 5 minutes.

19. Centrifuge and discard the supernate. Wash the fluoride with 5 ml H_2O, centrifuge and discard the supernate.

20. Add 3 ml sat'd H_3BO_3 to the precipitate, stir well and add 3 ml HNO_3. Stir to obtain a clear solution. Add 20 ml H_2O. Stir well and make basic with 6 N NaOH.

21. Centrifuge, discard the supernate, wash the hydroxide with 15 ml H_2O, centrifuge and discard the supernate.

22. Dissolve the final hydroxide in a minimum of 6 N HCl and continue with the Separation of Y from other Rare Earths.

b. Decontamination Procedures

1. Barium

(a) To the Ba carrier (about 20 mg) in a volume of 5 to 6 ml (in a 40 ml heavy-wall centrifuge cone), add the activity, stir thoroughly and let stand for ten minutes. Add 20 ml of yellow fuming HNO_3 to precipitate $Ba(NO_3)_2$ (note a). The solution is stirred while cooling in an ice bath for five minutes. Centrifuge, decant, and discard the supernate.

(b) Dissolve the precipitate completely in 2 ml of water (note b). Re-precipitate $Ba(NO_3)_2$ by adding 10 ml of fuming HNO_3 and cooling in an ice bath for 20 minutes with occasional stirring. Centrifuge the solution, decant, and discard the supernate. Dissolve the precipitate in 7 ml of water.

(c) Add 5 mg of Fe^{+++} carrier and precipitate $Fe(OH)_3$ by the addition of 2 ml of 6N NH_4OH while stirring (note c). Centrifuge and filter by gravity into a clean 40 ml centrifuge cone. Wash the precipitate by slurrying in 5 ml of H_2O containing 2 drops of NH_4OH. Centrifuge and remove the wash supernate with a transfer pipette. Combine this wash supernate with the supernate above and save. Discard the $Fe(OH)_3$ scavenge precipitate.

(d) Neutralize the combined supernates by the dropwise addition of 6N HNO_3, testing the acidity with short range pH paper. Add 1 ml of 6M HAc and 2 ml of 6M NH_4Ac. Heat the solution nearly to boiling and add 1 ml of 1.5M Na_2CrO_4, dropwise with stirring. Continue stirring the solution for one minute, then centrifuge. Save the supernate for step (e) of the strontium decontamination procedure. Wash the yellow $BaCrO_4$ with 10 ml of hot water. Centrifuge and discard the supernate. <u>The remaining steps in the procedure, including the final counting, must be finished within two hours to prevent error due to La-140 daughter contamination in the Ba precipitate.</u>

(e) Dissolve the $BaCrO_4$ in 2 ml of 6N HCl (note d). Add 15 ml of HCl ether reagent, note final PPT'n time, and stir for two minutes to coagulate the $BaCl_2$ and let stand for five minutes in an ice bath. Centrifuge the solution, decant, then discard the supernate.

(f) Dissolve the precipitate in a few drops of water (note b), and re-precipitate the $BaCl_2 \cdot H_2O$ by the addition of 15 ml of HCl ether reagent and stir the solution for two minutes. Allow the solution to cool in an ice bath for five minutes. Centrifuge, decant, and discard the supernate.

(g) Have ready a Whatman No. 42 paper disc which has been previously prepared by washing it with three 3 ml portions of water and three 3 ml portions of ethanol. Dry in an oven at 90^0-100^0 for 15 minutes, cool in a desiccator for ten minutes, and weigh after one minute on balance. Repeat this procedure until constant weight (± 0.1 mg) has been obtained.

(h) Dissolve the $BaCl_2$ in 10 ml of water. Heat the solution nearly to boiling and add 5 drops of 1.5M H_2SO_4. Continue heating for three minutes to coagulate the precipitate, then filter onto the prepared filter paper. Wash and dry the precipitate according to the filter paper treatment in step g. Weight as $BaSO_4$, mount, and count immediately (note e).

NOTES

a. At least 3 volumes of fuming nitric acid should be added to insure quantitative precipitation of $Ba(NO_3)_2$.

b. It may be necessary to add several additional drops of water to effect complete solution.

c. The addition of several drops of 1 per cent aerosol solution sometimes prevents excess creepage.

d. A precipitate of white $BaCl_2$ may appear but will dissolve with the addition of 0.5 ml of water.

e. The counting error due to growth of the La-140 daughter will become appreciable (approximately 3 per cent) after two hours from the $BaCrO_4$ precipitation.

2. Strontium

(a) To the Sr carrier (about 20 mg) in a volume of 5 to 6 ml (in a 40 ml heavy-wall centrifuge cone), add the activity and 20 mg of Ba carrier, stir thoroughly, and let stand for ten minutes. Add 20 ml of yellow fuming HNO_3 to precipitate $Ba(NO_3)_2$ and $Sr(NO_3)_2$ (note a). Cool the solution in an ice bath for 30 minutes with occasional stirring. Centrifuge at low speed for five minutes, decant, and discard the supernate.

(b) Dissolve the total precipitate with a minimum volume of water (about 2-3 ml) and repeat precipitation of the nitrates with fuming HNO_3 (note a). Centrifuge as above, decant and discard the supernate. Dissolve the precipitate in 7 ml of water.

(c) Add 5 mg of Fe^{+++} carrier and precipitate $Fe(OH)_3$ by the addition of 2 ml of 6N NH_4OH while stirring (note b). Centrifuge and decant the supernate through a Whatman No. 42 filter paper into a clean 40 ml centrifuge cone. Wash the precipitate by slurrying with 5 ml of H_2O containing 2 drops of NH_4OH. Centrifuge, remove the wash solution, and combine with the supernate above. Discard the $Fe(OH)_3$ scavenge precipitate.

(d) Neutralize the combined supernates by the dropwise addition of 6N HNO_3, testing the acidity with pH paper. Add 1 ml of 6M HAc and 2 ml of 6M NH_4Ac. Heat the solution nearly to boiling and add 1 ml of 1.5M Na_2CrO_4 dropwise with stirring. Continue stirring the solution for one minute, then centrifuge. Decant the supernate into a clean 40 ml centrifuge cone. Discard the $BaCrO_4$ precipitate.

(e) Heat the solution nearly to boiling and add 1 ml of $Ba(NO_3)_2$ solution (10 mg Ba/ml) dropwise with stirring. Continue stirring the solution one minute, then centrifuge. Decant the supernate through a Whatman No. 42 filter paper into a clean tube.

(f) Add 2 ml of NH_4OH, heat nearly to boiling, and add 5 ml of a saturated ammonium oxalate solution slowly with stirring. Continue stirring for two minutes. Centrifuge, decant and discard the supernate. The remaining steps in the procedure, including the final counting, should be finished within three hours to minimise the error due to Y-90 daughter in the Sr precipitate.

(g) Dissolve the SrC_2O_4 precipitate with 4 ml of 6N HNO_3. Add 15 ml of fuming HNO_3 (note c) stir well in an ice bath for 20 minutes with occasional stirring. Centrifuge, decant and discard the supernate.

(h) Dissolve the $Sr(NO_3)_2$ precipitate in 10 ml of water. Heat nearly to boiling and add 2 ml of 1M Na_2CO_3 solution slowly with stirring. Heat gently for several minutes and allow to cool in an ice bath for 10 minutes.

(i) Have ready a Whatman No. 42 filter disc which has been previously prepared by washing it with three 3 ml portions of H_2O and then with three 5 ml portions of ethanol. Dry in an oven at 90°-100°C for 10 minutes, cool in a desiccator for 10 minutes and weigh. Repeat this procedure until a constant weight (± 0.1 mg) has been obtained.

(j) Filter the $SrCO_3$ onto the prepared filter paper. Wash, dry, and weigh the precipitate according to the filter paper treatment in step i. Weigh as $SrCO_3$, mount, and count.

NOTES

a. At least three volumes of fuming HNO_3 should be added to insure the quantitative precipitation of the nitrates.

b. The addition of several drops of a 1 per cent aerosol solution will help prevent creepage.

c. Record the $Sr(NO_3)_2$ precipitation as the final precipitation time.

3. Molybdenum

(a) Add the solution containing the molybdenum activity to the molybdenum carrier (approximately 20 mg) and add sufficient HCl to make the solution 6N in HCl. Add 2 drops of bromine water and stir. Add one mg of Cs carrier and 1 mg of W carrier, stir thoroughly and let stand 10 minutes.

(b) Pour the solution through a Dowex-1-X10 short resin column (note a). Wash with 10 ml of 6N HCl, then with 10 ml of 0.1N HCl and finally with 10 ml of 3N NH_4OH.

(c) Elute the molybdenum off the column with 15 ml of 6N $NH_4C_2H_3O_2$ containing 5 ml of NH_4OH. Make the eluate just acid with HNO_3. Add 3 mg of Fe^{+3}, 5 mg of Zr^{+4}, 1 mg of Te^{+4} carrier, and 1 drop of 1M $NaNO_2$. Stir thoroughly. Make the solution at least pH 10 with NH_4OH.

(d) Stir thoroughly, centrifuge 2 minutes and filter through a Whatman No. 42 filter paper into a clean 40 ml centrifuge cone. Discard the filter paper and precipitate. Acidify the filtrate with HNO_3 to about pH 0 as indicated by short range pH paper. Add 7 drops of bromine water and 1 ml of saturated oxalic acid (note b). Stir and cool in an ice bath. Add 10 ml of filtered cupron reagent (2 per cent α-benzoin oxime in ethanol). Record final precipitation time. Stir very thoroughly again and centrifuge at high speed for 5 minutes. Decant carefully and discard the supernate. Wash the precipitate with 20 ml of 1N HNO_3. Centrifuge, decant and discard the supernate.

(e) Slurry the washed precipitate in 20 ml of 1N HNO_3 and filter through a 9 cm Whatman No. 42 filter paper very carefully in a Fisher Filtrator funnel under a slight vacuum (note c). Air dry by pulling air through filter paper and precipitate with the vacuum. Carefully fold and place the filter paper containing the precipitate in a coors No. 00 porcelain crucible. Tilt the cover

on the crucible and ignite in a muffle furnace for one hour at 525°C (note d). Record time taken out of furnace.

(f) Carefully grind the MoO_3 thoroughly in absolute ethanol using a glass rod. Filter through the stainless steel filter tower onto a 2.3 cm Whatman No. 42 filter disc previously prepared and tared. Wash with absolute ethanol. Dry in an oven at 90-100°C for 10 minutes, let cool 10 minutes in a desiccator and make a one minute timed weighing. Repeat drying, desiccating and weighing until agreement is within 0.1 mg. Mount, but do not count until 18 hours after furnace removal time (note e).

NOTES

a. The resin column is a short tubulated test tube of 12 mm I.D. and 85 mm long filled to a depth of about 1-1/4 inches with Dowex A.G. 1-X10, 50-100 mesh anion exchange resin. A glass wool plug is placed at the bottom and on top to contain the resin. The resin is pre-conditioned by washing with 10 ml of concentrated HCl. Discard this wash.

b. The oxalate ion complexes any niobium present, thus preventing its co-precipitation with molybdenum.

c. If too high a vacuum is pulled, the filter paper may tear and molybdenum will be lost.

d. The furnace should be calibrated and the temperature should not exceed approximately 525°C, as molybdenum can be volatilised by excessively high temperatures.

e. After 18 hours the daughter Tc^{99} will be in equilibrium with Mo^{99} and the sample may be counted.

4. Ruthenium

(a) To the solution containing the ruthenium activity in a volume of no more than 5 ml (in the ruthenium distillation apparatus), add Ru carrier in the desired amount. Add 25 ml of HNO_3. With the still head in place, boil until only a few ml of solution remain in the flask and add 3 ml of HCl.

(b) Have prepared a 40 ml centrifuge cone containing 10 ml of freshly prepared 12N NaOH in an ice bath. A system to provide a positive air flow through the apparatus during distillation must be utilized (note a). To the sample in the distillation flask, add 10 mgs of I^- carrier, then add 3 ml of HNO_3 and 4 ml of $HClO_4$. The ground glass connection should be wet with the $HClO_4$ to aid in sealing the apparatus. Connect the still head and start the air flowing through the system (about two bubbles per second). Heat the solution in the flask gently with a micro burner flame to begin the distillation. The distillation is continued until the dense white fumes of $HClO_4$ have passed over for one minute after all the Ru has been distilled (note b).
Caution: Considerable frothing may occur prior to the appearance of $HClO_4$. A sponge, or damp cloth, should be kept on hand to cool the flask in case the frothing becomes too violent.

(c) Let the distillate stand for 15 minutes, centrifuge and discard any residue. Adjust the pH of the distillate to 8, using dilute HCl or NaOH as needed. Add 3 ml of ethanol and stir well. Heat the solution until the precipitate coagulates (do not boil), centrifuge and discard the supernate.

(d) Dissolve the precipitate with 3 ml of HCl by allowing to stand a minimum period of three hours (note c). Check for complete dissolution of the precipitate by centrifuging. Transfer to a 125 ml flask and

dilute with 10 ml of water. Add powdered magnesium metal (note d) in small portions, swirling after each addition. Add the magnesium at a rate which will maintain a constant evolution of hydrogen until an excess of the powder has been added (when the HCl has been depleted the magnesium will cease to react and is then in excess). Boil the mixture until the Ru coagulates (note e). Cool the solution, and add 5 ml of HCl cautiously. Add 1 drop of 1 per cent aerosol solution, and boil the solution for two minutes. Transfer the material to a 40 ml cone and centrifuge. Decant and discard the supernate. Wash the Ru metal two times with water, centrifuging, decanting, and discarding the wash water each time.

(e) Have ready a weighed Whatman No. 42 filter paper disc which has been treated with three 5 ml portions each of water and ethanol. Dry the paper for ten minutes at 90-100°C, cool ten minutes in a desiccator, and weigh. Repeat this procedure until a constant weight has been obtained.

(f) Slurry with water and filter the precipitate through the filtration apparatus onto the weighed filter paper. Use the washing procedure and weighing procedure employed in preparing the filter paper, (note f). Weight as Ru metal (gravimetric factor 1.0). Mount and count.

NOTES

a. If a steady compressed air system is not available, a positive air flow may be obtained by displacing air in a bottle connected to the distillation apparatus by siphoning water from a higher second bottle.

b. The extremely dark coloring due to Ru will leave the distillation flask and appear in the NaOH.

c. It has been found that the subsequent reduction of Ru to the metal proceeds faster and more smoothly when the oxides are allowed to dissolve slowly at room temperature. However, if the long waiting period cannot be arranged, the solution may be heated gently to dissolve the oxides.

d. The magnesium must be residue free after dissolution with HCl. Test the magnesium by dissolving a gram in HCl.

e. If coagulation does not occur, or if the solution remains dark (indicating incomplete reduction), add HCl until the excess magnesium dissolves, then add 1 ml excess. Repeat the magnesium reduction.

f. After cooling in a desiccator, weigh the Ru precipitate without delay. Ru is somewhat hygroscopic.

5. Cadmium

(a) Add the solution containing the cadmium activity to the cadmium carrier (approximately 30 mg) in a 40 ml glass centrifuge tube. Adjust the volume to 10 ml with water. Adjust the solution to pH=7 with 6 N NH_4OH. Precipitate CdS with H_2S gas.

(b) Centrifuge and discard the supernate. Wash the CdS with 5 ml of water containing 2 drops of saturated NH_4NO_3 reagent. Centrifuge and discard the wash.

(c) Dissolve the precipitate with 1 ml of conc. HBr and boil. Add 0.5 ml of HNO_2 and boil (Note a). Cool and add 10 ml of conc. HBr.

(d) Elute the solution through a short column of Dowex AG1-X10 (50-100 mesh) resin (Note b). Wash the column with: 10 ml conc. HBr; and 10 ml of H_2O. Discard the washes.

(e) Place a clean 40 ml centrifuge tube under the column and elute the cadmium from the column with 20 ml of 6 N NH_4OH.

(f) To the eluate from Step e add 5 ml of NH_4OH and 5 mg of Fe^{+3} carrier. Stir and centrifuge for two minutes. Decant the supernate into a clean 40 ml centrifuge tube and elute through a short column of Dowex AG50-X12 resin (50-100 mesh) (Note c). Discard the $Fe(OH)_3$ precipitate.

(g) Wash the column with: 10 ml of 6 N NH_4OH; 10 ml of 1 N NH_4OH; and 15 ml of H_2O. Discard the washes.

(h) While Step g is in progress prepare 2.3 cm Whatman No. 42 filter discs by washing with distilled water and then with absolute ethanol. Dry at 90 - 100° C for 10 minutes, cool in a desiccator for 10 minutes and make a 1 minute timed weighing. Repeat drying, desiccating, and weighing to constant weight (±0.1 mg).

(i) Place a clean 40 ml centrifuge tube under the column and elute the cadmium from the column with 20 ml of 1 M HBr.

(j) To the eluate from Step i add 2 ml of 3 N NH_4Cl and adjust the pH to 8 with the dropwise addition of 6 N NH_4OH. Add

3 ml of 1.5 M $(NH_4)_2HPO_4$ and digest 15 minutes on a 80° C hot water bath. Maintain the pH of the solution at 8 with the addition of 6 N NH_4OH or 6 N HCl throughout the digestion.

(k) Filter the precipitate through the stainless steel filter tower onto the prepared paper disc. Wash with 5 ml portions of water. Allow the precipitate to air dry by suction. Dry in an oven at 90 - 100°C for 10 minutes, cool in a desiccator 10 minutes and make a one minute timed weighing. Repeat drying, desiccating and weighing until agreement is within ± 0.1 mg.

(l) Mount and allow 16 hours after final precipitation for the In^{115} daughter to grow into equilibrium before counting.

NOTES

a. If H_2S is not completely removed CdS will form in Step e.

b. The anion resin column is a short tubulated test tube of 12 mm I. D. and 85 mm long filled to a depth of about 1 inch with Dowex AG1-X10 (50-100 mesh) anion exchange resin. A glass wool plug is placed at the bottom and on top to contain the resin. The resin is pre-conditioned by washing with 10 ml of conc. HBr. Discard this wash.

c. The short cation resin column is the same as in Note b except the resin used is Dowex AG50-X12, 50 - 100 mesh cation resin. The resin is pre-conditioned by washing with 10 ml of 6 N NH_4OH. Discard this wash.

6. Cesium

(a) To the Cs carrier (about 20 mg) in a volume less than 8 ml (in a 40 ml heavy-wall centrifuge cone) add the activity and stir thoroughly. Add enough HCl to make the solution approximately 6 N in HCl.

(b) Add 1 ml of a 0.12 M silicotungstic acid solution and stir thoroughly. Digest the solution in a hot water bath for five minutes with occasional stirring. Allow the solution to cool for several minutes, centrifuge, decant and discard the supernate. Wash the precipitate with 5 ml of 6 N HCl, centrifuge, decant and discard the wash solution.

(c) Add 1.5 ml of 6 N NaOH to the cesium silicotungstate precipitate and heat gently with stirring until a clear solution results. Add 10 ml of 6 N HCl and heat gently for five minutes (a canary yellow precipitate of silicotungstic acid should be present). Cool the mixtures and allow to stand for five minutes. Centrifuge and decant the supernate into a clean 40 ml glass centrifuge tube. Wash the precipitate with 5 ml of 6 N HCl, centrifuge and combine with the supernate. Discard the precipitate.

(d) Repeat Steps b and c.

(e) Prepare a Whatman No. 42 filter paper disc by washing it three times with 5 ml portions of ethanol. Dry in an oven at 90° - 100° C for ten minutes, cool ten minutes in a desiccator, and weigh. Repeat this procedure until constant weight (± 0.1 mg) has been obtained.

(f) To the final solution, add 1 ml of 10 percent chloroplatinic acid (H_2PtCl_6) solution and stir thoroughly. Allow to stand for several minutes. Centrifuge and discard the supernate. Wash with two 5 ml portions of 6 N HCl, centrifuging and discarding the supernate each time. Wash with two 5 ml portions of absolute ethanol, centrifuging and discarding the supernate each time. (Note a).

(g) Slurry the precipitate in absolute ethanol and transfer into a previously tared porcelain crucible. Evaporate off the ethanol under a heat lamp. Dry the crucible at 90° - 100° C for 10

minutes, cool and weigh. Repeat this procedure until constant weight is obtained (± 0.1 mg).

(h) Weigh into the crucible enough dry, inert cesium chloroplatinate to give a final precipitate thickness of 15 mg/cm^2 (this amount can be calculated from the area of the precipitation apparatus used in the final mounting).

(i) Slurry the mixture with absolute ethanol and mix thoroughly. Filter quantitatively by repeated ethanol rinses of the crucible through the previously prepared filter paper disc. Wash down any precipitate adhering to the sides of the tower. Allow the paper disc and precipitate to dry with suction before removing the tower.

(j) Dry in an oven and weigh as in Step e. The final precipitate is weighed as Cs_2PtCl_6. The sample is mounted and counted.

NOTES

a. If the sample is to be counted on a gamma counter, skip steps g and h and proceed with step i.

7. Separation of Y from other Rare Earths

a. Resin Processing

Place about one pound of Dowex 50W-X4 (minus 400 mesh) analytical grade resin in a 2 liter beaker and add water to a height of four inches above the settled resin. Add 250 g dibasic ammonium citrate, stir well with an electric stirrer and adjust the pH to six with pH paper using 6 N NH_4OH. Stir for one hour with the electric stirrer.

Let stand 30 minutes, siphon off the fines and discard them. Again add water to a height of four inches above the resin, stir and let stand 30 minutes. Siphon off the fines as before. Repeat this process several more times until the supernate is quite clear.

Cover the graded resin with water and store for use.

b. Preparation of the Eluting Agent

Prepare about 2 liters of 0.5 M A-hydroxyisobutyric acid (AHIB) for each duplicate run. Dissolve 52 g AHIB in water, add one gram phenol and dilute to one liter.

About 1400 ml of 0.5 M AHIB at pH 3.10 and about 600 ml at pH 4.20 is used for each duplicate run. Make the pH adjustments with a Beckman Model G pH meter which has been calibrated with a BKH Standard pH 4.00 buffer solution.

c. Preparation and Conditioning of the Rare Earth Columns

1. Decontamination - Previously used R. E. columns are decontaminated by flushing several times with a boiling mixture of Versene and Alconox in water followed by several rinses with boiling distilled water.

2. Filling Columns with Processed Resin - Fill column half full with water. Take a small ball of pyrex wool (~1/2" diam.) which has been fluffed out to disorient the fibers and push plug to bottom of column

working air bubbles out as it goes down. Run a little water out of the stopcocks and check for air bubbles. Mark the column at a height 65 cm above the plug and add a water slurry of the resin while the water drips out of the stopcock. Don't allow the resin to go dry. Add resin till the settled resin is at the mark taking care to have a level surface and no resin particles on the glass walls above.

3. Conditioning the Resin Columns - The columns are conditioned with 0.5 M AHIB at pH 3.10. About two column volumes of this elutriant are allowed to pass through the columns under pressure (6.50 inches Hg) at room temperature. The columns may be conditioned overnight if necessary. After the columns have been conditioned, wipe the stopcock tips dry and coat with Desicote.

d. Loading of Rare Earth Samples onto Resin Column

1. The R. E. mixture (in 6 N HCl) is boiled down to a volume of calculated 1/2 ml. A few additional drops of 6 N HCl is added during boiling to prevent the R.E. from hydrolysing.

2. Let the R. E. solution cool to room temperature and then load onto the top of the resin column in small increments by transfer pipette, taking care that the tip of the pipette touches only a space about 1/8 inch above the resin bed. At no time should the level of liquid in the column touch the top of the resin bed. A loose fitting cork around the transfer pipette which will fit into the burette top aids in steadying the pipette during loading.

3. Wash the container 4 times with 1/2 ml portions of 0.5 M AHIB, pH 3.10, each wash being added onto the resin column in small increments.

4. The insides of the column above the resin bed is then washed down with several mls of 0.5 M AHIB, pH 3.10. These washes should be allowed to trickle down the insides of the column as slowly as possible so as not to disturb the top of the resin bed.

5. Fill the column to the very brim with 0.5 M AHIB (pH 3.10) and connect to the reservoir containing the elutriant. Wire all connections. All air bubbles must be removed.

e. Start of Elution

1. With all clamps, stopcocks, etc., shut off open pressure valve letting pressure to reducing valve. Adjust reducing valve slowly allowing pressure to build up to about 6.5 in. Hg with a small air leakage through the water bubbler.

2. Open in turn the remaining clamps and stopcocks. Check for air bubbles in liquid lines. Double check to see that the clamps are released and that the pH 4.2 sol'n. is able to flow into pH 3.1 sol'n. Start the magnetic stirrer.

3. Record time and date as "On Column Time."

4. The temperature of the system is kept at room temperature (25 to 30° C.).

5. The pressure is adjusted to obtain a flow rate of 26 - 29 ml/hr.

f. Collecting Fractions

1. The effluent is caught in 16 x 150 mm rimmed test tubes containing 2 ml of saturated oxalic acid.

2. Set the tubes on a turntable timed for 30 minute intervals. Check to make sure that the turntable controls are set to turn in the desired direction and the timer switch is "on". Check back later to see it is actually turning in the right direction.

3. Continue collecting until the Y band which elutes first is eluted.

g. Record Keeping

1. Resin - type, mesh, lot number

2. Resin bed - length and diameter

3. Elutrient - brand, concentration, volumes made up
4. Conditioning - length of time, volume through each, pressure.
5. On Column Time
6. Pressure - frequent observations of pressure and time and date.
7. Elution Data.

h. Selecting Fractions

1. Locating Peaks - A series of tubes containing a R. E. fraction will show a gradual increase in precipitate up to maximum, followed by a sharp decrease. The tube containing this maximum of precipitate is called the peak and its appearance is logged as final precipitation time.

2. The first and last tubes of a fraction are safeguarded against tailing of a previous fraction and early break-through of a subsequent fraction. The idea is to obtain enough precipitate for fairly good yields and yet provide margins on each side of a peak. If the shape of the elution peak is flat as judged by amounts of precipitate more tubes may be discarded on either side.

i. Mounting the Y Sample

1. Transfer the YC_2O_4 quantitatively to 40 ml centrifuge cone, centrifuging and discarding supernate.

2. Wash 3 times with water, 2 times with ethanol and filter through a stainless steel filter tower onto Whatman No. 42 filter paper. Use quantitative transferral technique.

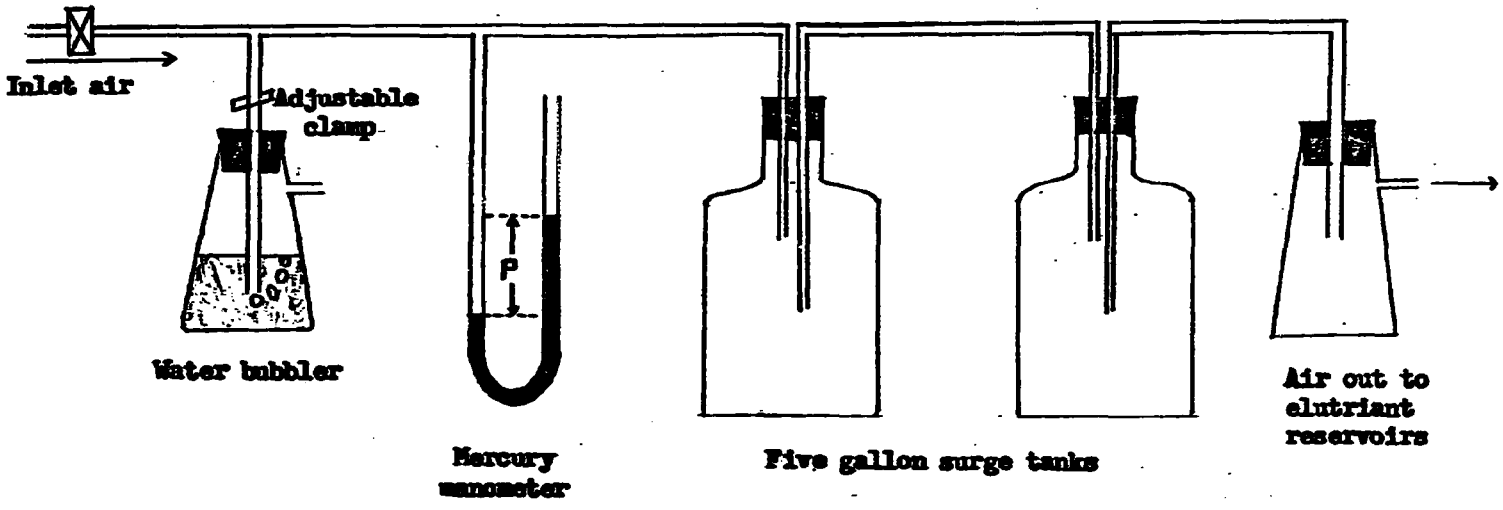
3. Ignite for one hour at 800° C.

4. Cool crucible, moisten the oxide with methanol and grind to a fine powder. Filter onto a tared No. 42 Whatman paper quantitatively.

5. Dry in oven at 90 - 100° C for 10 minutes.

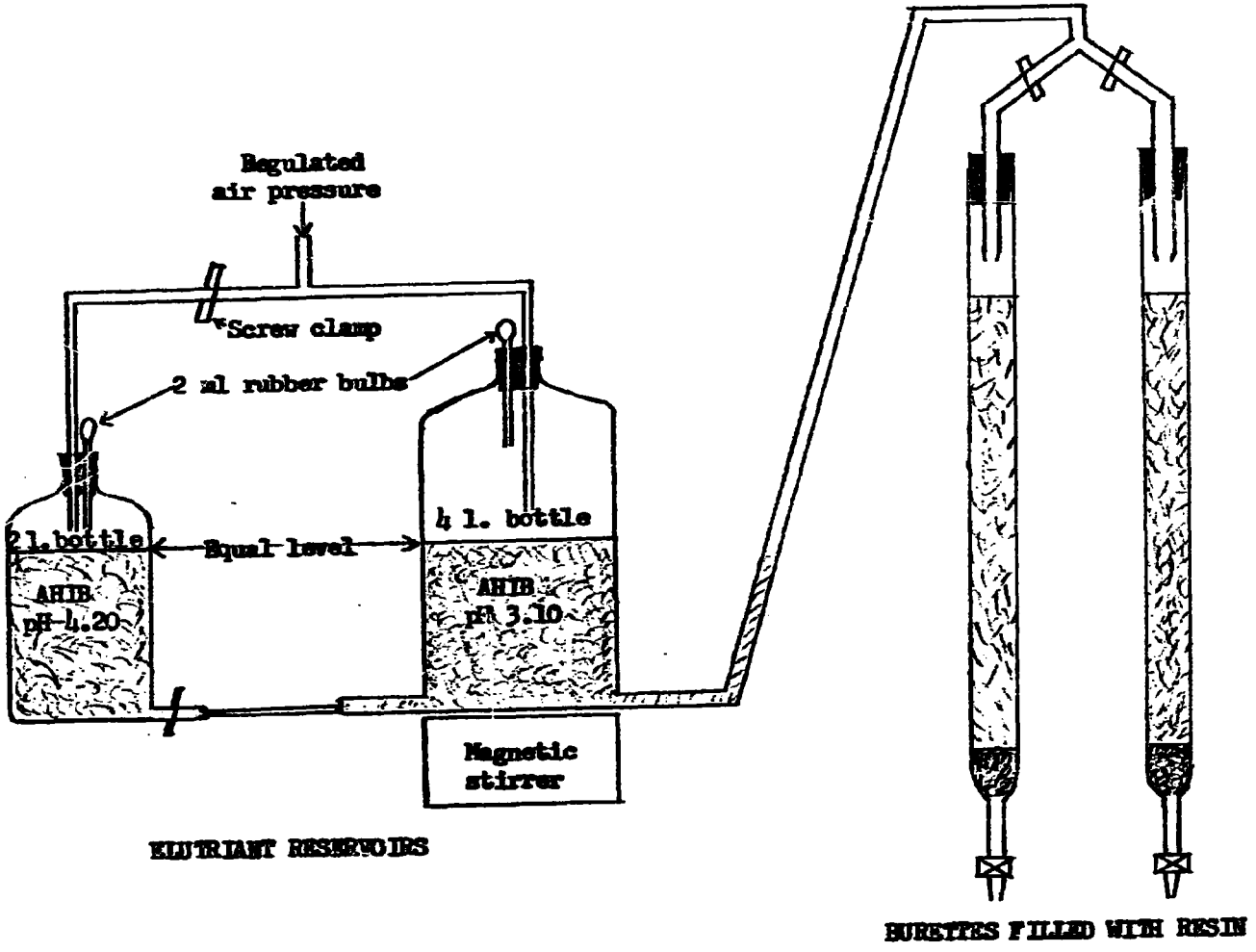
6. Cool in disiccator 10 minutes and take timed one minute weighing.
7. Repeat drying and weighing to constant weight ($\pm$ 0.1 mg).
8. Mount (brass planchet, pliofilm cover) and count.

FIGURE 1. AIR PRESSURE REGULATION



Note: Tygon and glass tubing used throughout.
All Tygon-glass connections wired on.
All rubber stoppers wired or clamped.

FIGURE 2. RARE EARTH COLUMN APPARATUS



Note: Tygon and glass tubing used throughout.
All Tygon-glass connections wired on.
All rubber stoppers wired or clamped.

8. Neptunium

(a) Add 3 mg of Fe^{+3} to the acid solution containing the neptunium activity. Precipitate $Fe(OH)_3$ by addition of excess NH_4OH and digest for a few minutes on a hot water bath. Centrifuge and wash the precipitate with 5 mls. 0.1 N NH_4OH. Centrifuge and discard the wash.

(b) Dissolve the $Fe(OH)_3$ with 3 drops HNO_3 and transfer to a 50 ml lusteroid tube with water washes. Make the total volume about 10 ml. Add 20 mg $NaHSO_3$ and heat the solution on a 75° water bath for 10 minutes. Add 5 mg La^{+3} and stir well. Add 10 drops HF and continue to heat for a few minutes. Cool, centrifuge and discard the supernate. Wash the LaF_3 with one ml 1 N HCl-1 N HF. Centrifuge and discard the wash solution.

(c) Slurry the LaF_3 in one ml saturated H_3BO_3 and heat a few minutes on a hot water bath. Stir and add one ml HCl. Stir and add one ml H_2O. Continue to heat for a few minutes until a clear solution is obtained. Transfer to a 40 ml glass cone with water washes.

(d) Make the solution ammoniacal with NH_4OH to precipitate $La(OH)_3$. Digest the hydroxide for five minutes on a 75° water bath. Centrifuge and discard the supernate. Wash the $La(OH)_3$ with five ml 0.1 N NH_4OH. Centrifuge and discard the wash solution.

(e) Dissolve the $La(OH)_3$ in one ml HCl. (Note A) Add 25 mg NH_4I. Cool in an ice bath and saturate with HCl gas. Allow the solution to come to room temperature and transfer to a prepared Dowex AG 1-X8 (100-200 mesh) column of one inch length. Elute through 15 ml HCl containing 50 mg NH_4I in 5 ml portions. Wash through 5 ml HCl. Elute the Neptunium into a 40 ml glass centrifuge cone with ten two ml portions of 1 N HCl-0.1 N HF.

(f) To the eluate from step e add 5 mg. Fe^{+3}. Make the solution basic with excess NH_4OH. Digest for 5 minutes on an 85° C. water bath. Centrifuge and discard the supernate. Wash the $Fe(OH)_3$ with 5 ml water containing 2 drops NH_4OH. Centrifuge and discard the wash.

(g) Dissolve the $Fe(OH)_3$ in 5 ml 1.5 HCl. Add one ml saturated $NH_2OH \cdot HCl$ and heat the solution 10 minutes on an 85° C water bath. Add another ml saturated $NH_2OH \cdot HCl$. Cool and add 5 ml benzene-0.4 M TTA. Stir the solution 3 minutes with a mechanical stirrer. Centrifuge for one minute to separate the layers. Withdraw the TTA phase with a transfer pipette and transfer to a clean, dry 40 ml glass centrifuge cone. Repeat the TTA extraction twice more and combine the benzene-TTA phases.

(h) Wash the benzene phase with 5 mls of 1.5 N HCl by stirring for two minutes with a mechanical stirrer. (Note B) Centrifuge and withdraw the aqueous layer. Repeat once and discard the washes.

(i) Back extract the neptunium from the TTA-benzene with 5 ml 8 N HNO_3. Stir for one to two minutes. Centrifuge and transfer the HNO_3 phase to a 50 ml beaker containing 2 mls $HClO_4$. Repeat the back extraction twice.

(j) Evaporate the solution just to dryness. Do not bake. Add one ml HCl and evaporate to dryness. Continue with steps two through four of the Heavy Element Electroplating Procedure.

(k) At the end of the plating period, turn off the stirring motor and with the current still on fill the cell with water. Remove the cell from the plating apparatus and immediately pour the solution into the 50 ml beaker previously used for evaporation in steps nine and ten. (Note C) Rinse the cell three times with 5 mls of water and combine the washes with the plating supernate.

(l) Dismantle the plating cell and remove the platinum disc. Rinse the disc with ethanol and ignite to red heat. Cool and mount for quick yield count.

NOTES

a. If instructions say "Repeat flouride", dilute sample to ten mls with water and repeat steps two through four. Do not add more La^{+3}.
b. Use 1.5N HCl standardized by titration.
c. The removal of the plating solution and rinsing of the cell must be done as rapidly as possible.

ELECTROPLATING PROCEDURE

1. Evaporate the solution containing the heavy element tracer and activity to approximately 1 ml. Add 1 ml HNO_3 and 1 ml HCl. Evaporate to wet dryness. Repeat the HNO_3-HCl treatment twice. (Repetition of HCl-HNO_3 treatment is not necessary for the plating of uranium.)
2. Pick up the sample in 1 ml HCl and take to dryness. Do not bake. Rotate the beaker to insure complete dryness. Add 2 ml HCl, boil to 1 ml and transfer to a prepared electroplating cell. Rinse the beaker with two 1/2 ml HCl washes and one 1/2 ml water wash. Transfer each wash to the plating cell. The platinum disc and anode must be freed of any grease film by rinsing several times with acetone and alcohol. Write the sample identification on the back of the disc. Ignite to red heat in a Fisher burner flame. The electroplating cell must be clean and free of any foreign material. Check for leakage before use. Keep the plating solution at minimum volume during this transfer and also during the following titration.
3. Add 1 drop methyl red indicator. Add NH_4OH dropwise until the indicator shows the solution to be basic (yellow). Add 2N HCl dropwise until the solution is just acid. Add 1 drop in excess.
4. Place the sample on a Sargent-Sloman electroplater. Adjust the anode to approximately 1/4 inch above the platinum disc. Plate for 10 minutes at a starting current of 2.5 amps and about 5 volts. The current may fluctuate during the plating period. Check occasionally and adjust the current to maintain 2.5 amps throughout the plating period.
5. At the end of the electroplating period, add 1 ml NH_4OH. Stir for 15 seconds. Turn off the current and stirrer and remove the anode from the plating solution.
6. Immediately transfer the plating solution into the beaker used for evaporation. Rinse the inside of the plating cell 3 times with water washes. Combine the washes with the plating solution.
7. Dismantle the plating cell and remove the platinum disc. Rinse with alcohol and ignite the disc to red heat. Cool and check the yield on an alpha counter. Place the sample in a lined and labeled tin box and submit for counting analysis.

5. Particle Analysis

Various methods of analyzing particles are available. In general, the specific method used depends upon the history of the sample, specific activity of the particles and what data is required from the particle investigation.

The following is a discussion of the procedures which are most likely to be applicable to analyzing particles from the Tory reactor.

a. Stir-off of the Activity from Filter Papers

Take a section of the filter paper of such size as to contain about 10^{9} fissions and cut into pieces of about 1/4" square. Place the pieces in a 40 ml centrifuge tube. Add 10 ml of n-amyl acetate to the tube and stir for 1/2 to 1 minute, at moderate speed, with a mechanical stirrer using a perforated glass tube as a stirring rod. Allow the fibers to settle for a few seconds and decant the amyl acetate through a double layer of 320 mesh wire cloth into a second 40 ml centrifuge tube.

b. Preparation of Collodion Films

Add 20 - 25 ml of Collodion Flexible, U. S. P., to the amyl acetate and stir well with a glass rod; do not mix air bubbles into the mixture as their presence will interfere with any microscopic examination which may be made. Place a few drops of silicone oil on a light glass plate and wipe well with a non-linting paper tissue. This coats the glass with a thin film of silicone to prevent adherence of the collodion film.

Pour a portion of the collodian-amyl acetate mixture onto the glass plate so that it covers an area of about 25 sq. inches. Dry for a few minutes under a 250 watt infrared heat lamp. (The heat lamp should be at least two feet distance to avoid formation of bubbles in the collodion.) Continue adding the collodion in increments to increase the thickness of the final completed film but keep the area to about 25 sq. inches.

Thoroughly dry the film under the lamp; four or five hours may be required. Remove the lamp and allow the film to stand for several hours. Remove the film from the glass plate using a razor blade to lift the edge of the film. Staple the film in a manila envelope to keep it held taut.

c. Radioautography of the Collodion Film

Drill a 1/4" hole in a piece of 1/8" sheet lead. Place the sheet lead over the end of a Tracerlab Lab Monitor Geiger tube shield so that the 1/4" hole is centrally located.

Take readings of the activity levels in the collodion film letting the lead sheet collimator rest on the film. Take readings as counts per minute and multiply by three to obtain the approximate activity level in terms of CPM/cm^2.

The following activity levels and exposure times have been found to give satisfactory radioautographs when Kodak Blue Brand Medical X-ray Film is used.

Activity Level CPM/cm^2	Exposure Time hours
1000	48
3000 - 5000	16
12000	5
20000	3

Staple an 8" x 10" Kodak Medical X-ray film over the collodion films mounted in the manila folder and punch a code for later rematching. Place the assembly in a Kodak X-ray Exposure Holder, weight lightly to get good contact between the collodion casts and film and expose for the proper time.

Remove the film and develop according to the following schedule.

Develop -- 6 min. at 20° C
- 5 min. at 23° C
- 4-1/2 min at 25° C

Stop Bath - 30 seconds
Fixer - 15 minutes
Wash - One hour
Allow the film to air dry.

d. Punch-out of Particles

Examine the radioautograph and circle the larger, round, black spots. If a single spot is not found the activity is not likely to be a single particle.

Rematch the radioautograph and collodion films. Staple securely in position. Punch out the marked spots. A punch with a 3/16" diameter hole is placed over the marked spot and then tapped with a hammer. The collodion circle is separated and the punch outs examined under a microscope to insure that the spot was caused by a large particle rather than an aggregation of minute particles.

e. Particle Dispersal Method Modification

If the filter paper is of low activity, the following outlined method may be used to isolate particles.

The filter paper is stapled to an x-ray film and exposed for a length of time determined by the activity of the paper. The film is developed and rematched with the paper. The spots on the film are punched out, using a 3/16" diameter punch and placed on a microscope slide. A drop of ethyl acetate-collodion solution is added to the punch-outs and the fibers are dispersed over a one sq. inch area using a pair of needles. The slides are then oven dried. The slides are exposed with x-ray film, using an aluminum jig to match the film and slide. The films are developed, rematched, the dark spot located under a microscope and the area under the spot scanned for the particle. The size and color of the particle are recorded and the particle transferred to a cover glass containing a drop of ethyl acetate which serves to wash away any adhering

collodion. The cover glass is then counted in a Tracerlab SC-16 2-π flow counter or 100 channel gamma analyzer and the specific activity calculated relative to the volume of the particle according to the following equation

$$(N_{\beta,\gamma,\alpha}) = \frac{(dpm)}{\frac{\pi}{6} D^3}$$

f. Radiochemical Analysis of Selected Particles

After isolating, determination of physical properties and specific activity determinations, the particles may be radiochemically analyzed either singly or in groups by dissolving in hydrofluoric-nitric acids, adding the suitable carriers and tracers and performing the appropriate separation, decontamination, and counting for the isotopes of interest as described in other sections of the appendix.

6. Counting Procedures

a. Barium-140

Direct count on a MEW (methane end window) counter to 2 percent accuracy immediately. If count rate is below 65 cpm, count the sample on a GE-14 low background counter. Sample must be counted within three hours of the final precipitation time.

b. Strontium 89 - 90

Direct count Sr sample on a MEW counter to 2 percent accuracy within three hours after the final precipitation time. Direct count the milked Y-90 daughter sample then decay daily until dead. If count rates are less than 100 cpm count samples on GE-14 low background counters.

c. Cesium 136 - 137

For Cs^{136} (12.9 day half-life) decay twice daily on a P-20 counter until the sample has decayed to 3 percent of its initial activity. Also count on the gamma spectrometer.

d. Ruthenium 103 - 106

Take an absorption curve from 0 to 1.6 gram 1 cm^2 aluminum on the MEW counter.

e. Molybdenum-99

Direct count on a MEW counter to within 1 percent accuracy. Decay three times a week for five half-lives (2.75 day half-life.).

f. Cadmium-115

Direct count on a MEW counter after 16 hours from final precipitation time has elapsed. This allows the In^{115} daughter to reach equilibrium. Decay for a purity check.

g. Yttrium-91

Direct count on a MEW counter, then decay twice a week for four weeks.

h. Neptunium-239

Direct count through a 4.75 mg/cm^2 aluminum absorber then decay twice daily on a MEW counter until four points have been taken. Change counting frequency to three times a week and decay until the sample is counting only 3 percent of its initial activity or until the counting rate starts to increase. Alpha count for yield determination.

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Unclassified Report

Methods for quantitative analyses of effluent samples from direct cycle air-cooled nuclear reactors are presented. Methods have been developed for analyses of beryllium and uranium at 10^{-9} gram levels in various collection media, and the level of beryllium and uranium present in the unexposed media has been determined. Separation and decontamination schemes have been developed for the detection of the following radionuclides: Mo^{99}, $Sr^{89,90}$, $Ru^{103,106}$, $Cs^{136,137}$, Ba^{140}, Y^{91}, Cd^{115} and Np^{239}. Collection media investigated included several filter papers.

1. Beryllium--analysis
2. Decontamination
3. Nucleons--analysis
4. Particle filters
5. Radiochemical analysis
6. Uranium
I. AFSC Project 1448
II. Contract AF 29(601)-2852
III. Tracerlab, Inc., Richmond, Calif
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