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RADIOCHEMICAL ANALYSIS OF DEBRIS FROM DISTANT NUCLEAR EXPLOSIONS.

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DEBRIS FROM THE 'HURRICANE' OPERATION

AN A.E.R.E. REPORT

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1. INTRODUCTION

Many radiochemical analyses of debris from distant nuclear explosions have been reported previously by British chemists (e.g. (Ref. 3)), but none on debris from an explosion of fissile material the constitution of which was certainly known. It has not been possible previously to sample the debris cloud at various distances from the origin in order to find the effects of distance and on the apparent ratios of the various nuclides present in the clouds. Again it has not been shown that different analysts working on the same active filter would find the same ratios. The "Hurricane" operation presented an opportunity of obtaining information of this kind, and the filter samples obtained from "Hurricane" clouds were analysed in a temporary radio-chemical laboratory set up in the Chemistry Dept. of the University of Melbourne, Australia. The team of chemists responsible for these analyses, authors of this report, constituted Team RC3 in the "Hurricane" experiment.

The results of the analyses are given in this report, but no attempt is made to discuss the results in relation to the general problem of interpetation; this will be done later when subsequent work can also be taken into account.

2. EXPERIMENTAL

Summary of Procedure

The cloud samples were standard British wire-gauze backed cylindrical paper filters through which air containing active dust had been filtered. No attempt was made to remove the particles from the paper; an area containing a convenient activity was cut out at random, carriers added and the whole subjected to a process which ensured complete destruction of the paper, and isotope exchange between carriers and activity. The carriers were then separated chemically along with the active isotopes, suitably mounted and β-counted. The isotopes of particular interest in this work were Fe^{59}, Sr^{89}, Zr^{95}, Mo^{99}, Ru^{103}, Ru^{106}, Ag^{111}, Pd^{112}, Cd^{115}, Ba^{140}, Ce^{141}, Ce^{143}, Ce^{144}, Np^{239}, Pr^{143}, Nd^{147}, Pm^{149}, Sm^{153}, and Eu^{156} and at the time no practicable simple analytical scheme including all these elements was known, hence two analytical schemes were employed both including Ba, Sr, Mo and Ag as reference elements and dividing the shorter lived elements more or less equally. Most of the purification schemes are based on those given in Ref. 2.

Analytical Procedures

Scheme 1. Sr, Ba, Mo, Ag, Pd, Ce, Np, Fe and rare earths

The active paper sample was wetted in a Pt dish with a solution of the carriers as nitrates, a known quantity of each (about 10 mgm. and approx. 500 counts/hr of Np^{237}) being used. The sample was then dried and partially carbonised under a heat lamp by the action of conc nitric acid, residual paper being destroyed in a muffle furnace at 500 ± 10°C for 1 hr. Any residual carbon was destroyed by evaporation with fuming HNO_3 (10 ml). Silica was then removed by evaporation with approx 1 ml. conc HF, followed by 1 ml conc HF + 1-2 mls conc H_2SO_4 until fumes of H_2SO_4 were evolved and all solid material appeared to be in solution in the H_2SO_4. The residue was cooled and diluted with 7-8 mls H_2O and the suspension digested for a few minutes in a steam bath, followed by cooling and standing for

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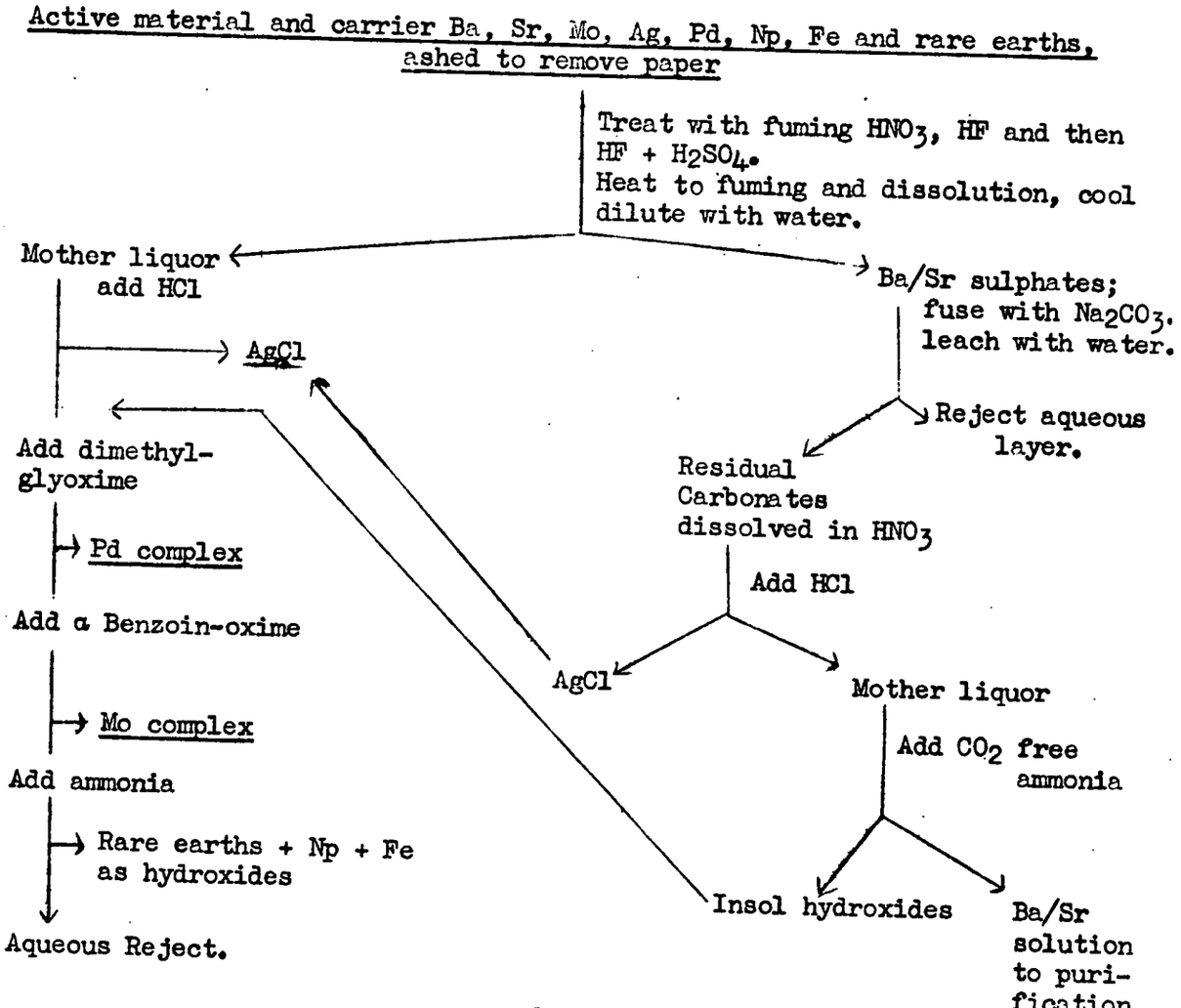
1-2 hrs. The mixed Ba/Sr sulphates were then centrifuged off, and fused with approx. 1g. of fusion mixture, or heated in a water bath for 15-20 mins with 5-10 mls saturated sodium carbonate solution; the carbonates thus formed were centrifuged free from an aqueous mother liquor which was discarded, and the residue dissolved in the minimum quantity of dil. HNO_3, diluted to 5 mls and any Ag removed as AgCl by addition of a few drops dil. HCl. The pptd AgCl was coagulated by warming, centrifuged off and reserved for treatment with the main Ag fraction. The mother liquor was made alkaline with CO_2 free ammonia solution and any ppte was centrifuged and returned to the main mother liquor from the Ba/Sr sulphate pptn. The alkaline solution containing Ba/Sr was reserved for purification. The main mother liquor was warmed and treated with a few drops dil. HCl to ppte AgCl which was removed by centrifuging and reserved for purification. The mother liquor was cooled and treated with 5 mls 2% dimethylglyoxime in alcohol, followed by drops of ammonia solution until a flocculent yellow ppte appeared. This ppte. of the Pd dimethylglyoximate was washed with 1-2 ml 0.5N HNO_3 and reserved for purification. The mother liquors and washings were treated with 3 mls of 2% alcoholic α-benzoin-oxime and the Mo complex thus pptd was removed by hard centrifuging, followed by washing with 5 mls 1N.HNO_3. The mother liquors and washings were now made strongly alkaline with ammonia to ppte rare earths, iron and neptunium together as hydroxides. The mother liquors from the hydroxides were rejected and on standing developed a crystalline organic ppte. If Sr had not been completely removed as described above it usually pptd. as sulphate from this mother liquor on long standing.

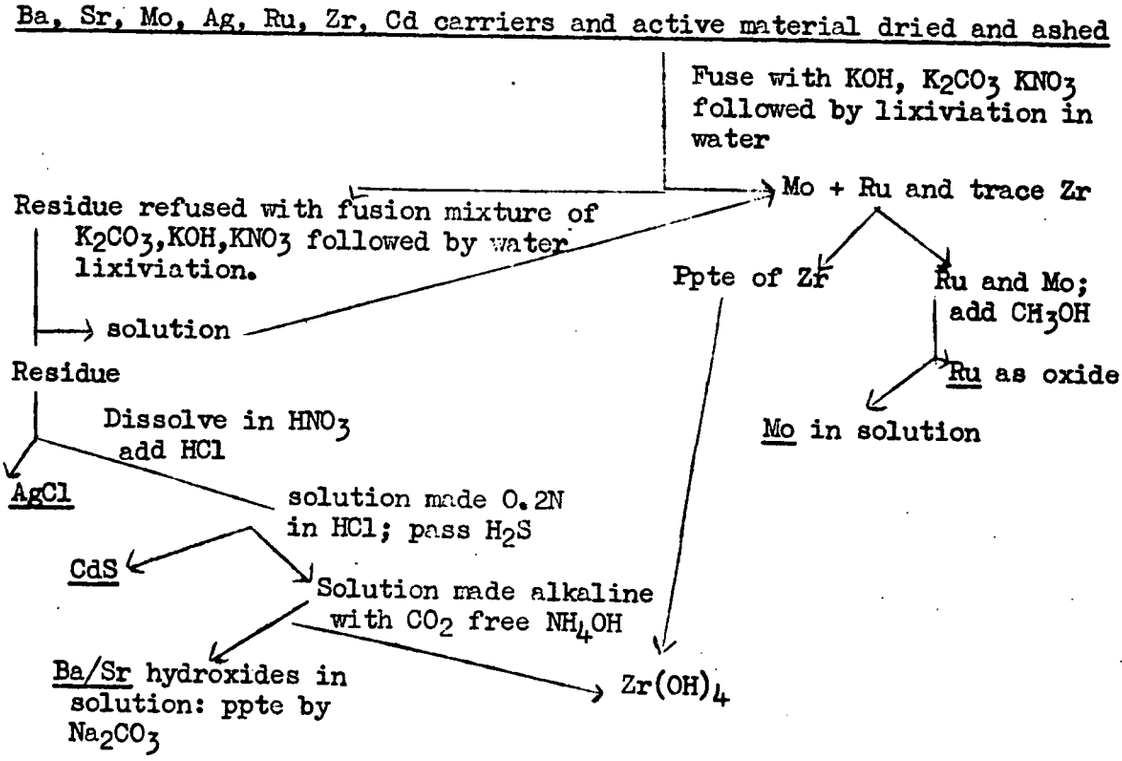
The outline of scheme 1 expressed diagrammatically is as follows.



Scheme 2. Sr, Ba, Mo, Ag, Ru, Zr and Cd

The active material was wetted with aqueous solutions of the carriers in known amounts (about 10 mgm), and the paper partly carbonised under a heat lamp. This process was quite rapid if done in a Ni crucible and no difficulty was experienced in completely ashing the sample at 500 ± 10°C for 30 mins. The residue was then fused in the Ni crucible with 3g. of a 1:1:1 mixture of K_2CO_3, KOH and KNO_3 for a few minutes. The cooled melt was lixiviated with water and centrifuged hard; the mother liquors were reserved for Mo and Ru analysis and the insolubles were dried under the heat lamp in the Ni crucible, and again fused with 0.5 g of the same fusion mixture. The cooled melt was lixiviated with water and the aqueous layer combined with the first for Mo and Ru analysis. $Zr(OH)_4$ occasionally pptd. in small quantities from this solution on standing. This was centrifuged off and added to the main Zr fraction.

The insoluble residue was boiled in the minimum quantity of dil. HNO_3 for dissolution. A slight greyish ppte containing Zr sometimes remained undissolved but this could be dissolved in a few drops of conc. H_2SO_4 and was added to the main Zr purification (see below). To the solution diluted to 7-8 mls with water, was added a few drops dil. HCl, the AgCl removed by centrifuging and the mother liquors made alkaline with dil. ammonia solution with stirring until a faint ppte just appeared. Sufficient HCl was then added with good stirring to redissolve this slight ppte, and to make the solution ~0.2N in acid. CdS was then pptd., by passing H_2S through the warmed supernate to ensure complete pptn of the Cd. The supernatant liquid was then boiled to removed H_2S and $Zr(OH)_4$ was pptd with a minimum of CO_2 free ammonia. The gelatinous ppte was spun down hard in the centrifuge and washed with a little dilute ammonia; Ba/Sr carbonates were then ppted. from mother liquor and washings by addition of sodium carbonate almost to saturation followed by standing over several hours in the cold. The mother liquor was rejected. Scheme 2 may be summarised diagrammatically as follows:-



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Purifications

Silver

AgCl pptes from the main separations were dissolved in a few drops conc. ammonia, the solution diluted to 5-10 mls and approx. 3 mgm. Fe as nitrate solution added drop by drop with vigorous stirring. The $Fe(OH)_3$ was centrifuged down hard and the supernatant liquor transferred to a clean tube. H_2S was passed to ppte. Ag_2S and this was centrifuged hard down and washed with water. The Ag_2S was dissolved in 1 ml conc. HNO_3 diluted to 5 mls and the solution made alkaline with ammonia. 3 mgms. Fe carrier was then added drop by drop with vigorous stirring and the whole cycle repeated. Finally the ammoniacal supernatant from the iron scavenging was made acid with HNO_3 and the whole solution warmed to boiling. 3-4 drops dil. HCl was then added and after approx. 30 secs. the whole solution was filtered by vacuum through a weighed filter paper, washed 3 times with 5 mls hot water, then 3 times with 5 mls. absolute ethanol. The combined paper and ppte. was dried at 110°C for 10 mins, stood in contact with the atmosphere for 2 hrs and weighed against a filter paper tare which had been in contact with the filter paper holding the ppte. at all times. The filter paper and ppte. were then stuck on to Al trays with perspex cement for counting, the ppte. being made firm on the paper by use of a solution containing approx. 0.5 mgm cellulose acetate. (This process is described later in more detail).

Molybdenum

The liquor containing the Mo from scheme 2 after removal of Ru (see below), was scavenged with 3 mgm (approx.) Fe carrier centrifuged hard down, the mother liquor was made acid with H_2SO_4 and 5 mls 2% alcoholic α-benzoin-oxime was added to ppte. the Mo complex, this was spun hard hard in the centrifuge. From this point Mo from both scheme 1 and scheme 2 were treated alike.

The Mo α-Benzoin-oxime complex was washed with 5 mls 1N HNO_3 and the ppte. was then dissolved by addition of 6N NaOH (approx. 2 mls.). The solution was diluted to 10 mls and 3 mgm Fe carrier was added with good stirring; the Fe ppte. was spun hard down and the Fe scavenging repeated. The mother liquor was then made approx. 2N in sulphuric acid and the Mo pptn. completed by addition of 2 mls. % alcoholic α-Benzoin-oxime. The complex was spun hard down, washed with 5 mls 1N HNO_3 and evaporated in a small beaker with 2 mls fuming HNO_3. When all liquid had evaporated, 5 mls fuming HNO_3 + 2 mls. 70% $HClO_4$ were added and heated continued almost to dryness. The residue was cooled and dissolved in 1 ml conc. ammonia and 10 mls water and the solution scavenged with 3 mgm Fe hydroxide. The mother liquor was examined to ensure that all $Fe(OH)_3$ was spun down and if not, further centrifuging was resorted to until the mother liquor could be removed cleanly from the $Fe(OH)_3$ ppte. The solution was made just acid with dil. H_2SO_4, buffered to a pH between 3.3 and 7.6 by addition of 2.0 mls 10% sodium acetate solution and the Mo ppted at boiling point as the complex of 8-hydroxyquinoline by addition of 3% dil. acetic acid solution of the reagent until no more complex forms and the supernatant is definitely yellow. The suspension was boiled 3 mins and filtered on to weighed papers, washed with hot water and alcohol and dried at 130°C for 10 mins followed by equilibration in air for 1 hr. The ppte. was then weighed and mounted as described later.

Barium

From scheme 1 the Ba/Sr fraction was in ammoniacal solution which was used direct; from scheme 2 the Ba/Sr carbonates were dissolved in

the minimum of dil. nitric acid and the solution boiled to remove CO_2; after cooling the solution was made ammoniacal with CO_2 free ammonia. The products from both schemes were at this point in the same form and were treated similarly. The ammoniacal solution was scavenged by addition of 3 mgm. Fe carrier with good stirring and the hydroxide ppte. spun hard down in the centrifuge. The mother liquor was evaporated to less than 1 ml and 7-8 mls of ether-HCl reagent was added, followed by cooling in ice for 10 mins; the crystalline ppte. was centrifuged down and the mother liquor reserved for Sr purification (see below). The ppte. was dissolved in less than 1 ml water containing 3 mgm La and 3 mgm. Sr (approx.), as hold back carriers, and 7-8 ml ether-HCl reagent were added followed by 10-20 mins. cooling in the ice bath. The ppte. was removed dissolved in minimum quantity of water and the $BaCl_2 2H_2O$ again pptd by addition of 7-8 mls ether-HCl reagent and cooling. The ppte. was then dissolved in 5 mls water, heated to boiling and $BaSO_4$ pptd. by addition of a few drops of dil. H_2SO_4. The ppte. was filtered through weighed papers, washed 3 times with hot water, containing 25% alcohol (5 mls) and 3 times with absolute ethanol (3 mls), and finally dried at 110°C for 10 mins. The ppte. and the filter paper were equilibrated in the atmosphere for 1 hour and then weighed and mounted.

Strontium

The Sr was contained in about 8 ml ether-HCl reagent from the Ba separation. The liquor was cooled in ice and 3 drops saturated $Ba(NO_3)_2$ in water added. After standing 10-20 mins the pptd $BaCl_2 2H_2O$ was centrifuged hard down and the process repeated. The mother liquor was evaporated to dryness the residue dissolved in 5 mls water a few drops 6N acetic acid and 2 mls 10% sodium acetate solution were added, and the pH was adjusted to 4.5 ± 0.5. 1 ml of 1.5N K_2CrO_4 solution was now added followed by 10 mgm Ba carrier drop by drop with good stirring. The stirring was maintained for a short while and the pptd $BaCrO_4$ centrifuged off. The Ba scavenging was repeated and the mother liquors were evaporated down to about 5 mls, and ethyl alcohol was added to make the solution 75% in alcohol. The pptd $SrCrO_4$ was filtered off, dissolved in 2 mls 6N HNO_3 and made up to 5 mls with 50% ethanol in water. The mixture was then heated to boiling point in a water bath and a few drops of sulphuric acid added to ppte. $SrSO_4$. The suspension was slowly cooled by standing at atmospheric temperatures, and was centrifuged down, washed with 50% aqueous alcohol, filtered on a tared filter paper, and treated in the same way as $BaSO_4$ (see above).

Rare earths, Neptunium and Iron

The mixed hydroxides were washed with water and centrifuged hard down and the wash discarded. The residue was dissolved in HCl to give about 10 mls of solution 8N in HCl. Ferric chloride was then removed by several extractions with isopropyl-ether (previously equilibrated with 8N HCl), and the solvent extracts combined, washed thoroughly with 8N HCl and the residue evaporated to dryness, dissolved in water and iron pptd as hydroxide with ammonia. The ppte was filtered on paper and mounted in the usual way for counting. No Fe activity could be detected.

The aqueous solution after solvent extraction was adjusted to 10 mls in a lusteroid tube, 5 mgm Zr carrier and 1 ml 1M $NH_2OH.HCl$ solution were added to ensure reduction of Np to IV-valent state, and the rare earths and Np were pptd by addition of 1 ml conc. HF. The ppt was centrifuged hard down, washed and dissolved in 1 ml conc. HNO_3 and 1 ml saturated H_3BO_3. 1 ml 2% $KMnO_4$ was added to the solution and the whole stood for 5 mins cold, with addition of more $KMnO_4$ if the

purple colour tended to fade. The solution was then diluted to 10 mls and the rare earths were pptd by addition of 10 mls. 10% NH_4F solution, centrifuged hard down and reserved for separation into the individual rare earths (see below). The mother liquor was decolourised by addition of sufficient $NH_2OH.HCl$ solution, 1.5 mgm Nd carrier added drop by drop with vigorous stirring over 20 mins and after standing 1 hr the ppte. was centrifuged hard down, dissolved in 1 ml conc. HNO_3 + 1 ml saturated H_3BO_3 solution and the whole cycle repeated, the Np being finally pptd on 1.0 mgm Nd and after washing, mounting on the standard Al tray by slurrying in water and drying under a heat lamp. This mounting would not interfere with the α-counting of the Np^{237} carrier.

The rare-earth fluoride ppte was dissolved in 1 ml conc. HNO_3 and 1 ml saturated H_3BO_3 diluted to 10 mls and the rare earths pptd as hydroxides with sufficient 6N NaOH. The washed ppte was then dissolved in the minimum of dilute HNO_3 slurried with 1 ml Zeo-Carb 225 cation-exchange resin and pipetted on to a 60 cm x 11 mm Zeo-Carb 225 column. The rare earths were then eluted one by one (except Eu and Sm collected together), by means of 5% ammonium citrate solution. During the elution the column was kept at 85°C by means of a heating jacket, and the resin of less than .05 mm particle diameter had been pretreated to remove particles which settled at less than 1 cm/min in water. The rare earths were detected in the column effluent by addition of oxalic acid to the 5 ml fractions collected, and the oxalates so ppted were centrifuged off washed with water, alcohol and ether, filtered and stood in air on the tared filter paper for 2 hrs before weighing. The ppte. on the paper was mounted as described later. In the case of Pm, it was usual to combine all the fractions between (Eu + Sm) and Nd and to treat the combined eluate with 10 mgm Nd followed by oxalic acid; this pptd. Pm on the Nd which was then treated in the same way as the other pptes. For reasons of convenience Eu and Sm fractions were combined and weighed and counted together.

Cadmium

The CdS was dissolved in 1 ml 6N HCl, diluted to 10 mls and 5 mgm Fe carrier were added, followed by ammonia until $Fe(OH)_3$ just began to ppte. The ppte was redissolved with 1 drop dil HCl, 0.5 ml 6M ammonium acetate added, and the whole solution heated in a water bath to ppte. basic ferric acetate which was centrifuged hard down and discarded. To the mother liquor was added 10 drops 6M HCl and H_2S gas was passed until all Cd ppted as sulphide; the ppte was centrifuged and the whole procedure repeated to this point. The ppte was then dissolved in 2 ml conc. HCl and the solution diluted to 10 mls, followed by addition of 5 mg Pd carrier. The solution was then heated and saturated with H_2S to ppt PdS which was centrifuged off and discarded. The acidity of the mother liquor was then reduced until CdS pptd on passing H_2S. The CdS was dissolved in a few drops of dil. HCl and H_2S removed by boiling; the diluted solution was then treated with ammonia until a ppte began to appear, this was dissolved by addition of a few drops of acetic acid followed by addition of 2 mls. 6M ammonium acetate. The solution was heated to 60°C and tested to ensure pH greater than 6, and the Cd pptd by addition of a slight excess of 2% alcoholic 8-hydroxy-quinoline. The solution was warmed to near boiling and filtered through tared paper, washed with hot water, alcohol and ether and dried for 10 mins at 130°C. The paper and ppte. were then equilibrated in the atmosphere for 1 hr and weighed and mounted as described later.

Palladium

The Pd complex of dimethylglyoxime was dissolved in 1 ml hot conc. HNO_3 the cooled solution diluted with 10 mls water, and 5 mgm Fe added

followed by excess ammonia. 10 mgm Ag carrier were then added together with just enough Iodide to ppte all the Ag. The ppte was centrifuged hard down and discarded. The pptn of Fe and Ag was repeated and the ppts discarded. The mother liquor was then made just acid with dil HCl and any AgCl centrifuged off. The cooled solution was treated with 5 mls 2% alcoholic dimethyl-glyoxime and the ppte stirred to coagulate. The Pd complex with dimethylglyoxime was then filtered through tared papers, washed with water and alcohol and dried at 110°C for 10 mins. After equilibrating in the atmosphere for 1 hr the ppte was weighed and the whole mounted as described later.

Zirconium

The hydroxide was dissolved in 1 ml conc. HF and diluted to 5 mls; 5 mgm of La were then added with good stirring and the LaF_3 ppte centrifuged hard down. A further 5 mgm La were then added and the suspension stirred and centrifuged hard down. The mother liquor was then treated with 1 ml saturated $Ba(NO_3)_2$ solution and the resultant ppte centrifuged off. The ppte was dissolved in 1 ml saturated H_3BO_3 and 1 ml conc. HNO_3 followed by addition of 3 mls water. A clear solution should result but sometimes a white ppte containing no Zr appeared which was removed by centrifuging. To the clear solution 1 ml conc. HF was added followed by 1 ml sat. $Ba(NO_3)_2$ solution and the whole allowed to stand until pptn. was complete. The ppte was removed and dissolved in saturated boric acid and conc. HNO_3 and the cycle repeated. Finally the $Ba\ Zr\ F_6$ was dissolved in 1 ml sat. H_3BO_3 and 1 ml conc. HCl and a clear solution obtained by dilution with 3 mls water. Ba was then pptd by addition of a few drops dil. H_2SO_4 and the pptd removed by centrifuging. To the mother liquor 2 mls 5% aqueous cupferron was added, the ppte centrifuged off, washed with 3-4 mls 1M HCl containing cupferron reagent and finally ignited to ZrO_2 in a silica crucible. The crushed ZrO_2 was mounted on paper and weighed, the whole being mounted for counting as described later. Alternatively the Zr was pptd after removing $BaSO_4$, by addition of an equal volume of saturated mandelic acid solution heating in the water bath for 30 mins and standing overnight. The pptd mandelate was centrifuged washed with alcohol and filtered through tared filter papers. After equilibrating for 2 hrs the ppte was weighed and mounted.

Ruthenium

This occurred in alkaline solution with Mo in scheme 2: 0.5g. KIO_4 were added to the alkaline solution followed by warming to complete the oxidation of the Ru. Sometimes a flocculent ppte was observed at this stage, which was Zr as the iodate; this was centrifuged out washed with a little water and added to the main Zr fraction. The mother liquors and washings were treated with 2 drops methyl alcohol and stirring and heating were continued until a black ppte settled out. This was centrifuged off and the mother liquors reserved for Mo purification. The black ppte was washed with approx. 3 mls 1N HNO_3 and the undissolved black oxide centrifuged down, dissolved in 5 mls 6N NaOH containing 0.5g KIO_4 with warming, the solution was cooled made acid with dil. H_2SO_4 and the RuO_4 extracted into CCl_4 by several washings of the aqueous phase with equal volumes of solvent. The CCl_4 combined extract was then shaken with 10 mls 6N HCl, the aqueous layer removed and the solvent shaken with more acid; this process was continued until the solvent layer contained only a faint pink colour or was colourless. The acid extracts were combined and evaporated to dryness in a beaker, the residue taken up in 5 mls water and treated with 3 mls 10% KI solution. On warming a black ppte of RuI_3 formed which was washed with water filtered through tared filter paper, washed with water, alcohol and ether and dried at 110 for 10 mins. After standing 1 hr in the atmosphere the ppte and papers were weighed and mounted.

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Mounting of samples for counting and transport

Since the experimental work described in this report was carried out in Melbourne, Victoria, Australia, it was necessary to transport samples for β-counting back to the U.K. for continued counting. A method of firmly mounting crystalline ppts on to flat Al counting trays was sought and found in the following procedure. The ppte was dried and weighed on a circular filter paper of 21 mm diameter, being in the form of a uniform circular microcrystalline pad of 18 mm diameter. This was attached to the paper by wetting pad and paper with a very dilute solution of cellulose acetate in acetone, so as to leave a maximum of 1 mgm cellulose acetate on the pad after evaporation of solvent. An aluminium counting tray (flat bottomed 1 inch lipped disc type), was then etched for 10 mins with 6N NaOH, and was washed thoroughly with water then acetone. Before the solvent could dry, a thin layer of 5% perspex in acetone solution (plasticised with tricresyl phosphate to 20% of the dry weight of perspex), was pipetted on to the tray and the paper disc with its adhering ppte. pad was floated on and pressed down round the edges so that the disc sank below the surface of the perspex solution, and was held thus until the greater part of the acetone had volatilised. On drying completely by standing in the atmosphere the ppte and paper were firmly welded to the counting tray. The counting tray was now fitted into the centre aperture of an Al plate and fixed by scotch-tape on the underside. The Al plate was an exact fit for insertion into standard Cook-type perspex fittings of a standard AERE lead castle, and when not counting the plate and sample tray were stored in grooved boxes fitted with lids. For transport to the U.K., the sample trays were covered with Al foil held in place by scotch-tape adhering to the Al plate, the plates being held firmly in the grooved boxes. This packing successfully withstood the vibration and pressure changes of the journey to AERE no samples being disturbed at all.

Counting of Samples

Samples mounted as described above were counted in standard AERE lead castles equipped with Cook-type perspex fittings. The detector was a thin windowed GM tube (type EHM2 window thickness = ~ 2 milligrams/cm^2 mica), connected via a 1014 type probe unit to 200 type scalers. The anode was maintained at known high voltage by means of a power unit type 1007 and potentiometer 1007. For those samples which were still active on arrival in the U.K. the absorption curves were obtained using an automatic absorber changer counter type 1112A.

The counters were set up and allowed to count natural background during the night hours; everyday a standard β-source was counted for 15 mins at about 3000 c/mins, the standard being U_3O_8 covered by 15 mgm Al foil and the whole stuck to Al counting trays of the same size as used in this work; periodic checks were made to ensure the absence of hysteresis in counting rate in the GM tubes used. These precautions enabled abnormalities in electronic equipment to be detected quickly, and before a day's counting programme had started. In reducing the counting data for the plotting of decay curves, the background and standard counts were plotted against time and a smooth curve was fitted to each plot. Interpolation of the time of a given count into the fitted curves enabled the appropriate backgrounds and standard counts, to be estimated. In fact the variation of these curves from a flat straight line was very small in a given location (Australia or England), and corrections were necessary only for background. The probe units of all the counters were preset at 300 μs dead-time, and when the counting rate was greater than a few hundreds/min it was necessary to make allowance for this.

For any one analysis all the elements concerned were counted in one particular counter (i.e. one particular EHM.2 tube since no changes were made during the major part of all decays), thus the geometry of counting was identical for all samples of a given analysis for a given position in the castle. Further, since one analyst used one counter only, all samples from all analyses made by a given analyst were counted under identical geometry for given castle positions.

If the counting rate for a given sample was too low for convenience when counted on the top shelf of the counter, it was usual to count it hard up to the EHM2 tube using an Al plate to define the distance from the window. These two positions were referred to in note-books as the "low" and "high" positions respectively.

Reduction of counting data

The counting data as recorded in note-books were corrected for paralysis time, background and (if necessary) standard variation; the resultant net counts were plotted against time on semi-logarithmic paper to allow of rapid visual evaluation of the decay curves. For the computation of lines of best fit the procedure varied according as the decay was nominally of a single isotope or the multiple decay of several isotopes of one element.

In the former case using $\log_e$ counts/min and time as variables the straight line of best fit was determined from which the apparent half-life could be found; the best straight line was then fitted assuming the best known half-life and an extrapolated estimate of the activity of the time of explosion was thus made. The presence of an impurity "tail" in the decay curve strongly influenced the apparent half-life and thus an idea of the radiochemical purity of the samples was obtained, and also an estimate of the activity not much influenced by the low levels of impurity found in fact. The standard deviations of the estimates made, naturally reflect the effects of impurities, together with the uncertainty of the best known half-lives.

In the latter case determination of the half-lives of the individual components although formally possible, was not practicable, and accordingly the best known half-lives of the component activities were assumed for any given sample, and the component activities estimated for the time of explosion, by least squares fitting. Separate estimates were thus obtained for each component activity of a mixture, and in considering these it must be remembered that they are not statistically independent estimates, and also that it has not been possible to include the effects of uncertainty in the best known half-lives used for the curve fitting.

In the cases of elements of long half-lives it was usual to count the first part of the decay curve in the "low" position, and the rest in the "high" position. The last part of a decay curve obtained under these circumstances shows up the presence of impurities very markedly and occasionally it has been necessary to analyse the "high" part of the decay curve assuming the presence of a likely impurity. This kind of effect is most marked in Sr^{89} decay curves where the presence of Sr^{90} begins to show after the majority of Sr^{90} begins to show after the majority of Sr^{89} has decayed away. Likewise, Ba^{140} and La^{140} decay curves sometimes show the presence of small quantities of Sr^{89}.

Summary of Results

Tables I to XVIII of the Appendix give summaries of the results obtained on the four counters from various air filters, and the identities and origins of the individual filters are given in Table I below; they have been more accurately described in Ref. 1.

TABLE I

IDENTITIES OF FILTERS USED

Filter	Ref. No.		Sampling height, distance and base of sampling aircraft
A B	1000/3 marked 534L 1000/6 marked 413M	1,2,3,4	(10,000', approx. 500 miles, Broome (
C_1 C_3	TV5D M TV5C M	1,4 2,3	(10,000' approx. 2000 miles. (Townsville
D_1 D_3	415 M 417 L	1,4 2,3	(500'-2000' approx. 1000 miles, (Broome
C_2	TV6CM	4	10,000' approx. 2000 miles, Townsville
D_2	418 M	4	500'-2000' approx. 1000 miles, Broome
E_1 E_2	WIDM WIC M	1 2	(10000' approx. 3500 miles, Fiji (Islands.

In the Appendix table I-XVIII under each isotope we have given the chemical yield of the carrier through the separation and purifications as obtained by direct weighing or in the case of Np^{239}, by α-counting the Np^{237} used as carrier. The absorption corrections are those accounting for both external absorption due to the window of the counting tube, the air gap between window and sample, and also to self absorption and back-scattering effects due to the carrier pptes and the mountings. The absorption corrections cannot be determined exactly for any given isotope and mounting conditions, there being a large experimental variance in both the extrapolated external and internal absorption and back-scattering curves; hence the standard deviation of the combined absorption corrections has been stated as a coefficient of variation along with the correction. Not all these corrections have been determined experimentally, some have been estimated from data on similar pptes. emitting similar radiation, a practice which is valid in these cases where the experimental variance is high in any case.

The "count at origin" is the estimated count extrapolated back to the time of explosion by means of the fitted curves, and the standard deviation quoted is that due to scatter of the points round the curve, and also to the uncertainty in the best known half-life used to estimate the slope (Ref. 6).

The "Corrected count at Origin" is the "count at Origin" corrected for chemical yield and with the absorption corrections applied; corrections due to decay systematics have also been applied e.g. Ba^{140} count actually consists of Ba^{140} and La^{140} in secular equilibrium, this has been corrected by dividing by 2.16. In the case of Cd^{115} a factor of 1/1.44 has been applied to correct for the conversion electrons of In^{115} daughter which are detected by the counter. In the case of Np^{239} the appropriate absorption corrections have been applied after determination empirically by counting known activities under the same conditions. Finally "Relative atoms at Origin" have been determined by multiplying the corrected count at origin by the half-life in days, and these values have then been used to calculate the ratios to Ba^{140}, which are set out in Table II. Throughout these tables the standard deviation of the figure concerned is given as a coefficient of variation; also the

difference between the "high" and "low" positions in counting have been preserved. This is because the self absorption and back-scattering corrections for the "High" counting position are not known accurately; they have been assumed to differ from the "Low" position corrections by a constant factor for all elements, a reasonable assumption on the basis of the work of Refs. 4 and 5 using relatively hard β-emitters, but uncertain in the case of soft β-radiation. Hence the figures given under "L" in Table II can be considered as estimates of absolute activities; those under "H" can be directly compared, but absolute values should be deduced by reference to the "L" values on the same sample.

Table III has been constructed as an indication of the agreement found between the three main ratios obtained in the first analyses on Filters A and B using different analytical methods.

Table IV has been constructed by taking mean values of the ratios to Ba^{140} of certain of the elements; in general the values are those obtained from the "L" counting position, but in the case of filter E they are all from the "H" position. Effectively this table compares the ratios obtained at various distances averaged over all other possible variables, and to illustrate the large overall effect which has operated on all the filters i.e. at all distances, the thermal fission ratios for Pu^{239} have been given for comparison. The data on Pu^{239} was obtained from Ref. 6, some of the ratios being interpolated from a curvilinear log (chain yield) V. mass number plot. In no instance was activity attributable to Fe^{59} found in the mounted samples from the analyses, and for this reason no mentioned of this element will be found in any of the Tables.

TABLE III

ATOM RATIOS TO Ba^{140} AT THE TIME OF EXPLOSION COMPARED FOR DIFFERENT ANALYTICAL METHODS

	Fusion Method			Wet ashing method		
	Sr^{89}/Ba^{140}	Mo^{99}/Ba^{140}	Ag^{111}/Ba^{140}	Sr^{89}/Ba^{140}	Mo^{99}/Ba^{140}	Ag^{111}/Ba^{140}
Filter A	.858 1.037 .947	.0444 .0505 .0474	.00438 .00498 .00468	1.041 .930 .985	.0457 .0468 .0462	.00467 .00476 .00472
Filter B	.757 1.097 .927	.0422 .0547 .0484	.00399 .00531 .00465	1.025 .904 .965	.0483 .0462 .0473	.00485 .00466 .00475
MEAN	.937	.0480	.00467	.975	.0468	.00473

Coefficient of Variation between operators on same sample Sr^{89}/Ba^{140} = ± 15.43%
Mo^{99}/Ba^{140} = ± 10.52%
Ag^{111}/Ba^{140} = ± 11.02%

TABLE IV

MEAN VALUES OF THE RATIOS TO Ba^{140} COMPARED FOR DIFFERENT DISTANCES FROM ORIGIN

Element	Filters A & B 500mls high cloud	Filters C 2000mls high cloud	Filters D 1000mls low cloud	Filters E 3500mls high cloud	Pu^{239} Thermal fission (Ref. 6)
Ba^{140}	1.0	1.0	1.0	1.0	1.0
Sr^{89}	.956	.843	.918	.734	.33
Mo^{99}	.0473	.0489	.0745	.035	1.13
Ag^{111}	.00470	.00487	.00633	.00544	.05
Ru^{103}	.127	.176	.133	.110	1.03
Ru^{106}	.163	.166	.163	.135	.87
Y^{91}	.499	.544	.534	.592	.52
Nd^{147}	.0108	.0124	.0192	.0174	.37
Pr^{143}	.0613	.667	.098	.0843	.95
Np^{239}	.555	1.057	1.22	-	-

3. DISCUSSION

(a) Analysis

The chemical procedures used in this work were very similar to and based on the experience of the work described in Ref. 3. There was one analytical problem outstanding which it was hoped to settle in this work, that is whether significant differences in the atom ratios were obtained using the fusion method (Scheme 2), for the destruction of paper and isotope exchange in the samples, as opposed to the wet ashing method (Scheme 1). In fact it is possible to compare only those analyses on Filters A and B (Run 1 in Table II), for the common ratios Sr^{89}/Ba^{40}, Mo^{99}/Ba^{149}, Ag^{111}/Ba^{140}, in all other analyses the activity levels available and laboratory accidents combined to make it impossible to give duplicated values on a given filter by a given method. The samples taken for analysis in the case of the Filters A and B were all removed from a small area of filter and it is reasonable to assume that the activity on each sample from a filter was the same. Examination of Table III which is derived from Table II will show that the mean values of the ratios to Ba^{140} of the isotopes Sr^{89}, Mo^{99}, Ag^{111}, are not significantly different for the different methods of analysis: indeed the coefficients of variation within repeat determinations by different operators are not much larger than those quoted in Table II for single analyses, notwithstanding the use of different methods and the introduction of possible personal biases.

It seems reasonable to assert therefore, that the two schemes of analysis used in this work yielded the same values of the ratios as judged by the radiochemical methods used. This is important since in future work it would be preferable to investigate all elements of interest in one analysis, and it is unlikely that the other elements concerned would be more affected by the ashing and isotope exchange technique used than Mo and Ag.

(b) Apparent effect of distance

Observations made on the cloud originating from the "Hurricane" explosion showed that it split into two parts (Ref. 1). The low level cloud proceeded into the Indian Ocean roughly North-West, and the high level cloud was blown over Australia roughly eastwards. Filters A, B, C and E therefore form a series of samples taken on the same part of the cloud at different distances, and differences in the ratios obtained from these filters may be attributed to the effect of "fall-out" on the constitution of the cloud. Examination of Table IV will suggest that such an effect does exist. There is however, one large effect common to all the filters, namely the large depletion of certain elements compared with the known thermal fission yields for Pu^{239} (Ref. 6; some data obtained by linear interpolation of these figures), and the effect of distance is small compared with this depletion. There seems to be little doubt that this large overall effect must be attributed to the siting of the weapon under water.

(c) Distribution of Activity on the filters

Although certain ratios obtained on Filters A and B were used in the discussion (a) above, to give evidence that the different analytical schemes used in this work gave the same results, it was pointed out that the samples used were all taken from a small area of the filter. Later analyses were made on Filters A and B on larger samples of the filters which were removed from the Filters at distances from the original excisions: these analyses are reported in Table II as Run 2 under the appropriate heading. It will be observed that the ratios obtained in these runs were significantly different from those obtained in the

first runs, notably in the cases of the Sr^{89}/Ba^{140} and Ag^{111}/Ba^{140} ratios. Further it will be observed that the Ce^{141}/Ba^{140} and the Ce^{144}/Ba^{140} ratios found in all the analyses show large difference. These facts suggest that the activity on the filters was not uniform, even in the cases of Filters A and B, of high activity, and where the activity was contained in very minute particles (N. G. Stewart - Private Communication). More positive conclusions could have been drawn if all the elements of interest had been investigated on each sample analysed.

4. CONCLUSIONS

Since all analyses made on distant samples from the "Hurricane" explosion have shown great depletions in some elements with respect to Ba^{140}, it is desirable that similar analyses on the cloud of a weapon fixed above ground level should be obtained before general conclusions are drawn concerning the information obtainable from air-samples. Further information should also be sought on the effects of fall-out in altering the constitution of the cloud during travel, and the fact that activity collected on filters may well be non-uniform should be borne in mind. It would also be desirable to analyse for all elements of interest on each sample, in view of the possibility of non-uniform distribution on the filter.

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APPENDIX TABLE 1 - COUNTER 1. FILTER A. SUMMARY OF RESULTS

Isotope	Ba^{140}	Sr^{89}	Mo^{99}	Ag^{111}	Cd^{115}	Ru^{103}	Ru^{106}	
Chemical Yield	67.1%	14.75%	77.7%	79.1%	50.0%	60.1%	60.1%	
Absorption Correction	.805 ± 3.85%	.87 ± 2.52%	1.147 ± 4.72%	.763 ± 5.31%	0.822 ± 4.70%	3.04 ± 7.18%	.795 ± 4.49%	Refers to "Low" position in counting
Half life found	12.71 ± .03 days	51.1 ± .4 days	66.8 ± 0.27 hrs.	185.7 ± 2.4 hrs.	56.3 ± 1.1 hrs.	-	-	
Count at origin	20,700 ± 0.8%	400.1 ± 3.23%	1586 ± 3.96%	89.0 ± 3.0%	213.7 ± 10.5%	89.6 ± 2.1%	40.7 ± 3.12%	
Corrected count at origin	12,360 ± 3.94%	2,360 ± 4.10%	2,340 ± 6.16%	85.8 ± 6.10%	244 ± 11.50%	450.9 ± 7.48%	53.6 ± 5.47%	
Relative atoms at origin	147,100 ± 5.55%	126,260 ± 5.68%	6,533 ± 6.34%	644 ± 6.32%	549 ± 12.08%	17,946 ± 7.51%	19,564 ± 6.81%	
Best known half life	12.8 ± .05 days	53.5 ± 2.1 days	67 ± 1 hrs.	180 ± 3 hrs.	54 ± 2 hr.	39.8 ± .3 days	365 ± 15 days	
Half life found	-	57.9 ± .6 days	-	-	-	-	-	
Count at origin	196,500 ± 40.7%	3,029 ±10.5%	-	-	-	751.9 ± 3.61%	305.7 ± 1.21%	Refers to "High" position in counting
Corrected count at origin	117,350 ± 40.88%	17,865 ± 10.8%			- -	3,784 ± 8.01%	402.4 ± 4.65%	
Relative atoms at origin	1,397,000 ± 41.06%	955,778 ± 11.49%			-	150,603 ± 8.06%	146,876 ± 6.20%	

APPENDIX TABLE III COUNTER 1. FILTER C1. SUMMARY OF RESULTS

Isotope	Ba^{140}	Sr^{89}	Mo^{99}	Ag^{111}	Cd^{115}	Ru^{103}	Ru^{106}	Zr^{95}	
Chemical yield	24.9%	29.5%	63.6%	76.0%	41.0%	39.9%	39.9%	45.5%	
Absorption correction	.805 ± 3.8%	.84 ± 2.5%	1.147 ± 4.7%	.763 ± 5.31%	0.833 ± 4.7%	2.60 ± 7.18%	.814 ± 4.4%	1.834 ± 9.00%	
Half life found	-	-	-	-	-	-	-	-	
Count at origin	-	190.1 ± 1.2%	288.1 ± 3.2%	19.2 ± 5.77%	32.5 ± 10.2%	-	-	-	
Corrected count at origin	-	541 ± 2.8%	520 ± 5.73%	19.3 ± 7.8%	46.0 ± 11.2%	-	-	-	
Relative atoms at origin	-	28,944 ± 4.8%	1452 ± 5.92%	145 ± 8.02%	104 ± 11.8%	-	-	-	
Best known half life	12.8 ± 0.05 days	53.5 ± 2.1 days	67 ± 1 hrs.	180 ± 3 hrs.	54 ± 2 hrs.	39.8 ± 3 days	365 ± 15 days	65 ± 2 days	
Half-life found	12.69 ± .04 days	54.1 ± 0.3 days	66.1 ± .53 hrs.	179.0 ± 2.5 hrs.	57 ± 1.6 hrs.	-	-	73.0 ± 1.2 days	
Count at origin	10,910 ± 1.2%	1326 ± 6.7%	1996 ± 4.95%	133.1 ± 3.18%	268 ± 12.67%	132.0 ± 1.6%	43.7 ± 1.3%	31.2 ± 4.3%	
Corrected count at origin	17,636 ± 4.0%	3776 ± 7.16%	3600 ± 6.8%	134 ± 6.19%	378 ± 13.51%	860 ± 7.36%	89.2 ± 4.69%	126 ± 9.97%	
Relative atoms at origin	209,940 ± 5.6%	202,016 ± 8.2%	10,051 ± 7.02%	1005 ± 6.42%	851 ± 14.00%	34,228 ± 7.41%	32,560 ± 6.2%	8,190 ± 10.4%	

Refers to "Low" position in counting

Refers to "High" position in counting

APPENDIX TABLE IV COUNTER 1. FILTER D1 SUMMARY OF RESULTS

Isotope	Ba^{140}	Sr^{89}	Mo^{99}	Ag^{111}	Cd^{115}	Ru^{103}	Ru^{106}	Zr^{95}	
Chemical Yield	36.7%	31.2%	62.8%	71.3%	45.1%	54.2%	54.2%	54.8%	Refers to "Low" position in counting
Absorption correction	.805 ± 3.8%	.84 ± 2.52%	1.147 ± 4.7%	.763 ± 5.31%	0.822 ± 4.7%	2.893 ± 7.18%	.795 ± 4.49%	1.922 ± 9.00%	
Half life found	12.59 ± .06 days	50.5 ± .4 days	67 ± 0.96 hrs.	176.1 ± 3.1 hrs.	55 ± 1.6 hrs.	-	-	70.25 ± .95 days	
Count at origin	14,770 ± 1.2%	1293 ± 3.61%	2572 ± 4.42%	124.0 ± 3.38%	217.2 ± 13.2%	115.4 ± 2.82%	49.0 ± 3.45%	36.0 ± 2.20%	
Corrected count at origin	16,200 ± 4.06%	3481 ± 4.40%	4698 ± 6.47%	133 ± 6.29%	275 ± 14.01%	616 ± 7.7%	71.9 ± 5.66%	126 ± 9.26%	
Relative atoms at origin	192,840 ± 5.64%	186,234 ± 5.90%	13,120 ± 6.66%	998 ± 6.52%	619 ± 14.49%	24,517 ± 7.76%	26,240 ± 7.18%	8190 ± 9.76%	
Best known half life	12.8 ± 0.05 days	53.5 ± 2.1 days	67 ± 1 hrs.	180 ± 3 hrs.	54 ± 2 hrs.	39.8 ± 3 days	365 ± 15 days	65 ± 2 days	
Half-life found	-	57.7 ± .6 days	-	-	55.2 ± 3.4 hrs.	-	-	67.3 ± 1.3 days	Refers to "High" Position in counting
Count at origin	115,000 ± 3.36%	9747 ± 10.64%	17,990 ± 4.58%	909.5 ± 1.44%	1505 ± 10.7%	1189.3 ± 19.6%	348.1 ± 2.1%	304.5 ± 6.85%	
Correct count at origin	126,125 ± 5.11%	26,242 ± 10.9%	32,858 ± 6.58%	973 ± 5.50%	1905 ± 11.69%	6348 ± 20.87%	511 ± 4.96%	1068 ± 11.31%	
Relative atoms at origin	1,501,000 ± 6.42%	1,404,000 ± 11.63%	91,740 ± 6.78%	7298 ± 5.76%	4286 ± 12.26%	252,650 ± 20.88%	186,515 ± 6.68%	69,420 ± 11.72%	

APPENDIX TABLE V - COUNTER 1. FILTER E.1 SUMMARY OF RESULTS

Isotope	Ba^{140}	Sr^{89}	Ag^{111}	Ru^{103}	Ru^{106}	Y^{91}	Nd^{147}	Pr^{143}	Ce^{141}	Ce^{144}	
Absorption Correction	.805 ± 3.85%	.788 ± 2.52%	.763 ± 5.3%	3.149 ± 7.18%	.783 ± 4.49%	.844 ± 5.90%	.922 ± 5.08%	.825 ± 7.65%	1.29 ± 3.74%	1.014 ± 3.70%	These refer to the "High" position in counting
Chemical Yield	72.8%	59.9%	76.0%	66.7%	66.7%	70.3%	74.5%	70.1%	49.9%	49.9%	
Half life found	13.6 ± 2 days	51.6 ± 6 days	-	-	-	62.9 ± 7 days	-	14.07 ± .27 days	-	-	
Count at origin	1,332 ± 2.57%	93.8 ± 3.22%	7.24 ± 21.8%	5.16 ± 13.7%	2.78 ± 13.0%	86.4 ± 5.24%	13.1 ± 7.35%	46.3 ± 1.78%	55.8 ± 0.9%	1.25 ± 18%	
Corrected count at origin	737 ± 4.53%	123 ± 4.09%	7.27 ± 22.44%	24.4 ± 15.47%	3.26 ± 13.75%	103.7 ± 7.89%	16.2 ± 8.93%	54.5 ± 7.85%	144 ± 3.75%	1.27 ± 181.38%	
Relative atoms at origin	8,774 ± 5.98%	6,581 ± 5.66%	54.5 ± 22.50%	971 ± 15.49%	1,190 ± 14.35%	6,118 ± 8.58%	183 ± 9.10%	747 ± 7.88%	4,608 ± 4.90%	354 ± 18.44%	
Best known half life	12.8 ± 0.5 days	53.5 ± 2.1 days	180 ± 3 hours	39.8 ± 3 days	365 ± 15 days	59 ± 2 days	11.3 ± .02 days	13.7 ± 0.1 days	32 ± 1 days	294 ± 4 days	

APPENDIX TABLE IX COUNTER 2. FILTER D3. SUMMARY OF RESULTS

Isotope	Ba^{140}	Sr^{89}	Mo^{99}	Ag^{111}	Np^{239}	Y^{91}	Nd^{147}	Pr^{143}	Ce^{144}	Ce^{141}	
Chemical Yield	78.7%	23.7%	81.0%	45.1%	24.1%	77.1%	100%	88.5%	46.6%	46.6%	
Absorption Correction	.805 ± 3.85%	.872 ± 2.52%	1.147 ± 4.72%	.763 ± 5.31%	.838 ± 3.0%	.844 ± 5.90%	.922 ± 5.08%	.825 ± 7.65%	1.014 ± 3.7%	1.333 ± 3.74%	
Half-life Found	12.69 ± .04 days	51.04 ± 1.57 days	67.6 ± .64 hrs.	-	63.7 ± 2.1 hrs.	58.8 ± .013 days	-	13.43 ± .09 days	-	-	
Count at Origin	32,100 ±1.11%	767.8 ± 3.49%	3574 ± 5.90%	118.0 ± 2.7%	29500 ± 1%	1353 ± 4.00%	-	1070 ± 1.18%	16.6 ± 25.5%	987 ± 1.0%	Refers to "Low" position in counting
Correct count at Origin	16,417 ± 4.01%	2825 ± 4.30%	5061 ± 7.56%	200 ± 5.96%	102,577 ± 13.34%	1484 ± 7.13%	-	997 ± 7.74%	18.0 ± 25.77%	2824 ± 3.87%	
Relative atoms at origin	195,430 ± 5.60%	151,138 ± 5.82%	14130 ± 7.72%	1500 ± 6.20%	239,346 ± 13.81%	87,379 ± 7.90%	-	13659 ± 7.78%	5022 ± 25.81%	90,368 ± 5.0%	
Best known half-life	12.8 ± .05 days	53.5 ± 2.1 days	67 ± 1 hrs.	180 ± 3 hrs.	56 ± 2 hrs.	59 ± 2 days	11.3 ± 0.2 days	13.7 ± 0.1 days	279 ± 4 days	32 ± 1 days	
Half-life Found	14.34 ± .24 days	58.01 ± 6.32 days	-	179.6 ± 1.6 hrs.	-	59.68 ± 1.11 days	-	15.01 ± .25 days	-	-	
Count at Origin	244,500 ± 5.56%	5610 ± 10.23%	23,520 ± 5.27%	625.9 ± 2.98%	-	9477 ± 8.88%	2257 ± 1.88%	7767 ± 6.07%	181.4 ± 2.04%	7033 ± 1.03%	Refers to "High" position in counting
Corrected count at Origin	125,046 ± 6.76%	20,641 ± 10.54%	33,305 ± 7.07%	1,059 ± 6.09%	-	10,374 ± 10.66%	2,081 ± 5.42%	7,240 ± 9.77%	198 ± 4.13%	20,122 ± 3.88%	
Relative atoms at origin	1,488,000 ± 7.82%	1,104,000 ± 11.25%	92,988 ± 7.24%	7,943 ± 6.32%	-	612,066 ± 11.18%	23,515 ± 5.70%	99,188 ± 9.80%	55,242 ± 4.38%	643,904 ± 5.0%	

APPENDIX TABLE X - COUNTER 2 - FILTER E.2 - SUMMARY OF RESULTS

Isotope	Ba^{140}	Sr^{89}	Ag^{111}	Mo^{99}	Y^{91}	Pr^{143}	Nd^{147}	Ce^{141}
Chemical Yield	70.4%	53.6%	77.5%	74.3%	94.7%	77.7%	81.7%	12.0%
Absorption Correction	.805 ± 3.85%	.846 ± 2.52%	.763 ± 5.31%	1.147 ± 4.72%	.844 ± 5.90%	.825 ± 7.65	.922 ± 5.08%	1.205 ± 3.74%
Count at origin	1,468 ± 5.51%	85.0 ± 5.07%	6.3 ± 12.8%	80.9 ± 8.3%	92.3 ± 4.96%	57.3 ± 1.82%	10.94 ± 4.30%	17.4 ± 2.9%
Corrected count at origin	839 ± 6.72%	134 ± 5.66%	6.2 ± 13.86%	125 ± 9.55%	82.3 ± 7.71%	60.8 ± 7.86%	12.3 ± 6.99%	174 ± 4.73%
Relative atoms at origin	9,987 ± 7.76%	7,169 ± 6.88%	46.5 ± 13.96%	349 ± 9.67%	4,856 ± 8.42%	833 ± 7.90%	139 ± 7.22%	5,600 ± 5.60%
Best known half-life	12.8 ± .05 days	53.5 ± 2.1 days	180 ± 3 hrs.	67 ± 1 hrs.	59 ± 2 days	13.7 ± 0.1 days	11.3 ± 0.2 days	32 ± 1 days
Half-life found	14.3 ± 19 days	53.11 ± .42 days	287.1 ± 112.7 hrs.	64.8 ± 11.0 hrs.	57.4 ± .43 days	13.7 ± .33 days	9.35 ± .77 days	-

Refers to "High" position in counting

APPENDIX TABLE XI COUNTER 3. FILTER A. SUMMARY OF RESULTS

Isotope	Ba^{140}	Sr^{89}	Mo^{99}	Ag^{111}	Cd^{115}	Zr^{95}	Ru^{103}	Ru^{106}	
Chemical Yield	63.8%	35.3%	69.3%	73.5%	54.5%	39.5%	51.3%	51.3%	Refers to "Low" position in counting →
Absorption Correction	.805 ± 3.85%	.832 ± 2.52%	1.147 ± 4.72%	.763 ± 5.31%	.687 ± 4.70%	1.747 ± 9.00%	3.90 ± 7.18%	.783 ± 4.49%	
Half-life Found	12.96 ± .06 days	52.12 ± .35 days	66.6 ± .32 hrs.	176.8 ± 3.2 hrs.	63.6 ± 1.7 hrs.	74.9 ± 1.82 days	-	-	
Count at Origin	15230 ± 1.13%	939.7 ± 3.77%	1,250 ± 3.67%	73.0 ± 3.04%	200.7 ± 10.24%	16.86 ± 3.52%	40.26 ± 6.80%	39.00 ± 4.09%	
Correct count at origin	9,608 ± 4.01%	2,215 ± 4.53%	2,069 ± 5.98%	75.8 ± 6.02%	176 ± 11.26%	74.7 ± 9.66%	306 ± 9.89%	59.5 ± 6.07%	
Relative atoms at origin	114,400 ± 5.60%	118,502 ± 6.00%	5,777 ± 6.16%	569 ± 6.14%	396 ± 11.85%	4,856 ± 10.14%	12,179 ± 9.92%	21,718 ± 7.34%	
Best known half-life	12.8 ± .05 days	53.5 ± 2.1 days	67 ± 1 hrs.	180 ± 3 hrs.	54 ± 2 hrs.	65 ± 2 days	39.8 ± 3 days	365 ± 15 days	
Half-life Found	16.3 ± .4 days	57.08 ± .6 days	-	-	-	68.55 ± 1.24 days	-	-	← Refers to "High" position in counting
Count at Origin	107,503 ± 2.62%	6,966 ± 10.72%	-	-	-	175.9 ± 6.79%	455.6 ± 6.31%	231.4 ± 1.75%	
Corrected count at origin	67,822 ± 4.66%	16,418 ± 11.01%	-	-	-	778 ± 11.27%	3,464 ± 9.56%	353 ± 4.82%	
Relative atoms at origin	807,350 ± 6.08%	878,363 ± 11.69%	-	-	-	50,570 ± 11.68%	137,867 ± 9.50%	128,850 ± 6.34%	

APPENDIX TABLE XII COUNTER 3 FILTER B SUMMARY OF RESULTS

Isotope	Ba^{140}	Sr^{89}	Mo^{99}	Ag^{111}	Cd^{115}	Zr^{95}	Ru^{103}	Ru^{106}
Chemical Yield	33.4%	17.9%	73.8%	73.5%	52.5%	42.0%	58.5%	58.5%
Absorption Correction	.805 ± 3.85%	.861 ± 2.52%	1.147 ± 4.72%	.763 ± 5.31%	.694 ± 4.70%	1.775 ± 9.00%	4.193 ± 7.18%	.772 ± 4.49%
Half life found	13.77 ± 1.0 days	50.72 ± .66 days	67.2 ± 88 hrs.	176.9 ± 4.4 hrs.	63.5 ± 1.93 hrs.	-	-	-
Count at origin	10,930 ± 7.79%	464.7 ± 2.33%	1,371 ± 2.62%	74.22 ± 2.74%	187.4 ± 10.08%	17.35 ± 3.54%	56.6 ± 4.88%	42.8 ± 3.83%
Corrected count at origin	9,136 ± 8.69%	2,235 ± 3.47%	2,131 ± 5.40%	77.0 ± 5.95%	172 ± 11.78%	73.3 ± 9.67%	406 ± 8.60%	56.5 ± 5.90%
Relative atoms at origin	100,755 ± 9.53%	119,570 ± 5.24%	5,950 ± 5.62%	578 ± 6.22%	387 ± 12.35%	4,765 ± 10.15%	16,159 ± 8.96%	20,620 ± 7.18%
Best known half life	12.8 ± 0.05 days	53.5 ± 2.1 days	67 ± 1 hrs.	180 ± 3 hrs.	54 ± 2 hrs.	65 ± 2 days	39.8 ± 3 days	365 ± 15 days
Half life found	-	56.47 ± .48 days	-	-	-	68.74 ± .29 days	-	-
Count at origin	122,324 ± 3.17%	3,470 ± 9.64%	-	-	-	184.8 ± 6.09%	446.7 ± 10.78%	292.9 ± 2.04%
Corrected count at origin	59,036 ± 4.99%	16,691 ± 9.96%	-	-	-	781 ± 10.87%	3,202 ± 12.95%	387 ± 4.93%
Relative atoms at origin	702,740 ± 6.34%	892,969 ± 10.71%	-	-	-	50,765 ± 11.30%	127,400 ± 13.25%	141,255 ± 6.42%

Refers to "Low" position in counting

Refers to "High" position in counting

APPENDIX TABLE XIII COUNTER 3. FILTER C3. SUMMARY OF RESULTS

Isotope	Ba^{140}	Sr^{89}	Mo^{99}	Ag^{111}	Cd^{115}	Zr^{95}	Ru^{103}	Ru^{106}	
Chemical Yield	77.0%	41.5%	72.3%	65.9%	52.2%	45.4%	34.1%	34.1%	Refers to the "Low" position in counting
Absorption Correction	.805 ± 3.8%	.82 ± 2.5%	1.147 ± 4.7%	.763 ± 5.3%	.81 ± 4.7%	1.813 ± 9.0%	2.479 ± 7.1%	.828 ± 4.4%	
Half-life Found	13.33 ± .12 days	52.7 ± .41 days	67.7 ± 3.95 hours	-	-	-	-	-	
Count at Origin	3796 ± 8.6%	137.8 ± 3.7%	264.8 ± 4.8%	-	-	-	-	-	
Corrected count at origin	1985 ± 9.5%	272 ± 4.5%	420 ± 6.7%	-	-	-	-	-	
Relative atoms at origin	23,630 ± 10.2%	14,552 ± 5.9%	1,173 ± 6.9%	-	-	-	-	-	
Best known half-life	12.8 ± .05 days	53.5 ± 2.1 days	67 ± 1 hrs.	180 ± 3 hrs.	54 ± 2 hrs.	65 ± 2 days	39.8 ± 3 days	365 ± 15 days	
Half-life Found	18.05 ± .47 days	55.9 ± .82 days	65.3 ± 3.5 hrs.	180.1 ± 3.56 hrs.	68.2 ± 1.7% hrs.	64.9 ± 1.6 days	-	-	Refers to the "High" position in counting
Count at Origin	24,093 ± 3.5%	1,015 ± 10.1%	1,906 ± 5.7%	107.9 ± 3.5%	354.2 ± 14.1%	27.1 ± 4.3%	97.6 ± 1.9%	29.7 ± 2.4%	
Correct count at origin	12,594 ± 5.2%	2,006 ± 10.4%	3,024 ± 7.4%	125 ± 6.3%	384 ± 14.8%	108 ± 9.9%	710 ± 7.4%	72.1 ± 5.0%	
Relative atoms at origin	149,900 ± 6.5%	107,320 ± 11.1%	8,443 ± 7.5%	938 ± 6.5%	860 ± 15.3%	7,020 ± 10.4%	28,258 ± 7.4%	26,316 ± 6.5%	

APPENDIX TABLE XIV COUNTER 3. FILTER D3. SUMMARY OF RESULTS

Isotope	Ba^{140}	Sr^{89}	Mo^{99}	Ag^{111}	Cd^{115}	Zr^{95}	Ru^{103}	Ru^{106}	
Chemical Yield	62.8%	42.5%	85.2%	62.8%	50.3%	42.1%	31.4%	31.4%	Refers to the "Low" position in counting
Absorption Correction	.805 ± 3.85%	.819 ± 2.52%	1.147 ± 4.72%	.763 ± 5.31%	.813 ± 4.70%	1.777 ± 9.00%	2.424 ± 7.18%	.825 ± 4.49%	
Half-life Found	12.96 ± .08 days	53.4 ± .25 days	66.5 ± .66 hrs.	.179 ± 6.4 hrs.	61.3 ± 3.59 hrs.	-	-	-	
Count at origin	20,340 ± 1.24%	1,049 ± 3.63%	3,305 ± 5.63%	97.8 ± 2.64%	227.5 ± 11.9%	-	69.9 ± 3.65%	30.8 ± 4.76%	
Corrected count at origin	13,036 ± 4.04%	2,021 ± 4.42%	4,449 ± 7.35%	119 ± 5.93%	256 ± 12.83%	-	540 ± 8.05%	809 ± 6.54%	
Relative atoms at origin	155,180 ± 5.62%	108,120 ± 5.92%	12,422 ± 7.50%	893 ± 6.16%	576 ± 13.35%	-	21,492 ± 8.08%	29,529 ± 7.72%	
Best known half life	12.8 ± .05 days	53.5 ± 2.1 days	67 ± 1 hrs.	180 ± 3 hrs.	54 ± 2 hrs.	65 ± 2 days	39.8 ± 3 days	365 ± 15 days	
Half-life Found	15.8 ± .63 days	55.0 ± .5 days	-	170.4 ± 1.86 hrs.	57.2 ± 4.4 hrs.	68.9 ± .5 days	-	-	Refers to "High" position in counting
Count at origin	143,120 ± 8.51%	7,482 ± 9.48%	23,110 ± 4.54%	712 ± 4.03%	2,000 ± 20.0%	249.1 ± 4.91%	592.4 ± 8.87%	202.1 ± 1.33%	
Correct count at origin	91,729 ± 9.34%	14,418 ± 9.81%	31,111 ± 6.55%	865 ± 5.67%	2,245 ± 21.33%	756 ± 10.25%	4,573 ± 11.41%	531 ± 4.68%	
Relative atoms at origin	109,182 ± 12.6%	771,360 ± 13.6%	86,862 ± 6.72%	6,488 ± 6.88%	5051 ± 21.65%	49,140 ± 10.70%	182,000 ± 11.71%	193,815 ± 6.16%	

SI ARD

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S.R.O. HARWELL