

EUROPEAN ATOMIC ENERGY COMMUNITY - EURATOM

# NEUTRON ACTIVATION ANALYSIS OF ALUMINUM

### DETERMINATION OF GAMMA-EMITTING IMPURITIES WITH LONG HALF LIVES

by

F. GIRARDI, R. PIETRA

1963



Joint Nuclear Research Centre Ispra Establishment (Italy) Nuclear Chemistry Service

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# Neutron Activation Analysis of Aluminum

### Determination of Gamma-Emitting Impurities with Long Half Lives

FRANCESCO GIRARDI and ROMANO PIETRA Servizio Chimica Nucleare, Centro Comune di Ricerche Euratom, Ispra (Varese), Italy

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**PURE ALUMINUM** is frequently used in nuclear reactor technology for its low neutron absorption cross section and rapid decay of the induced activity. The presence of some impurities, even at the trace level, has a rather negative effect on the nuclear properties of aluminum. These impuri-

ties include fissile and fertile elements (uranium, thorium), elements with high neutron absorption cross section (boron, cadmium, and many rare earths), and elements giving, upon neutron activation, radioisotopes of long half life (cobalt, iron, chromium, scandium, cesium, etc.). The impurities that give rise to long-lived gamma activity are of special interest from the standpoint of radiation hazard involved when equipment must be irradiated for a long time and then extracted from the reactor and handled for further operation or disposal. The determination of these elements at the trace level is therefore of great importance in establishing the nuclear properties of a material proposed for use in reactor experiments. Neutron activation analysis shows an excellent sensitivity for all of these elements with the exception of boron, and it is therefore frequently employed for these analyses.

We have developed an analytical scheme for the above elements based on the following criteria: Most of the elements with an important influence on nuclear properties should be determined with a suitable sensitivity and precision; elements of minor significance should also be determined at least semiquantitatively, if present at unusually high levels; the operation time should be kept sufficiently low to permit the analysis of many samples in a reasonable Automation of the chemical time. separations and instrumental analysis should be used, whenever possible. Sequential separation schemes on many materials analyzed by neutron activation are available from the literature.

Blaedel, Olsen, and Buchanan (4)

have studied on synthetic mixtures of radioisotopes the possibilities of cation exchange for a group separation of metallic radioelements. The procedure is very rapid and it may be easily automated, but unfortunately for our purpose the greatest part of the elements of interest for reactor technology fall within one group.

Separation schemes on a large number of elements have been studied in neutron activation analysis of semiconductors (21) and water (5). Other schemes have been studied for the separation of individual fission products in health physics research (6, 8). Also in these cases the range of elements studied does not cover many of the elements of interest in the nuclear field.

Albert has developed the systematic analysis by neutron activation of high purity metals (aluminum, iron, zirconium); 49 elements are determined in a single sample. The first procedures were rather lengthy (2), but in his latest work only 2 working days are necessary for such a complete analysis (1).

Jervis and Mackintosh (11) have used ion exchange for determining impurities in reactor grade aluminum. Mackintosh (14) used radiochemical separations to study the distribution of impurities in the purification of aluminum by zone melting. Since none of the above separation schemes met our requirements, particularly that of rapidity, we have developed a new scheme.

The elements determined with our procedure are reported in Table I. The procedure covers the range of gamma-emitting isotopes with half lives greater than approximately 1 day.

Table I. Long-Lived Gamma-Emitting Impurities Determined in Neutron Activated Aluminum

Group	Element	Radio- isotope deter- mined	Half life	Gamma energy	Sensitivity, p.p.n.ª	Chem- ical yield, %
1	Hafnium	Hf <sup>181</sup>	46  d	0.13, 0.34, 0.48	$2.7 imes10^{-4}$	95
	Thorium	Pa233	27.4 d	0.31	$4.9 \times 10^{-4}$	99
2	Cobalt	$Co^{60}$	5.3 y	1.17, 1.33	$1.4 \times 10^{-3}$	96
	Uranium	$Np^{239}$	2.3 d	0.21, 0.28	$6.0 \times 10^{-4}$	98
3	Iron	Fea	45.1d	1.10, 1.29	0.5	100
	1 in Win a	Sn113 77-65	112 d	0.39	0.5	99
1	Cadmium	Calus	$\frac{240}{52}$	0.51, 1.12	$1.4 \times 10^{-2}$	99
3 6	Silver	0.0110 A.:.110	$\frac{1}{20}$	0.34, 0.32 0.45 0.69 0.99	$25 \times 10^{-4}$	98
0	Suver	Ag	210 a	0.40, 0.00, 0.00, 1.30, 1.30, 1.52	2.5 X 10 ·	99
7	Cesium	$C_{8}^{134}$	2.3 11	0.60 0.79	$1.2 \times 10^{-3}$	99
•	Rubidium	B 686	186d	1 08	$1.9 \times 10^{-3}$	98
8	Scandium	Sc.16	$\hat{85}$ $\hat{d}$	0.89.1.12	$6.7 \times 10^{-5}$	98
Ū.	Lanthanum	$\tilde{L}a^{140}$	40.2h	0.33, 0.49, 0.81, 1.60, 2.50	$1.1 \times 10^{-4}$	00
	Samarium	$\mathrm{Sm}^{153}$	47.0 h	0.07, Ó.10	$2.8  imes 10^{-5}$	
	Holmium	H0 <sup>166</sup>	27  d	0.08, 1.36, 1.53	$1.9 \times 10^{-4}$	
	Cerium	Ce <sup>141</sup>	32  d	0.145	$9.2 \times 10^{-3}$	99
	Neodimium	$Nd^{147}$	11.3 d	0.10, 0.52	$4.3  imes 10^{-3}$	95
	Terbium	$\mathrm{Tb}^{160}$	73.0 d	0.09, 0.20, 0.29, 0.41, 0.89, 0.96, 1.18, 1.27	$1.2 \times 10^{-4}$	
	Thulium	${ m Tm}^{170}$	127 d	0.08	$8.0 \times 10^{-5}$	
	Ytterbium	Yb <sup>169</sup>	$\overline{32}$ $\overline{d}$	0.11	$3.9 \times 10^{-4}$	
	Lutecium	$\mathrm{Lu}^{177}$	6.7 d	0.11, 0.21	$1.3 \times 10^{-5}$	
9	Chromium	$Cr^{51}$	27.8 d	0.32	$1.5 \times 10^{-3}$	97

<sup>a</sup> Sensitivity has been calculated as the quantity that gives 1000 disintegrations per minute in 0.2 gram of aluminum irradiated for 24 days in a neutron flux of  $2 \times 10^{13}$  neutrons per sq. cm. per second.

These isotopes show a residual activity when the shorter-lived Cu<sup>64</sup>, Ga<sup>72</sup>, and Na<sup>24</sup> have decayed. The Na<sup>24</sup> activity is produced by sodium impurities and by irradiation of the aluminum with neutrons of energy high enough to permit the reaction:

$$Al_{13}^{27} + n_0^1 \rightarrow Na_{11}^{24} + He_2^4$$

The determination of the elements shown in Table I is adequate for the prediction of the dose rates of radiation from aluminum components irradiated for long periods in nuclear reactors. Uranium and thorium are included for the calculation of fission products production during irradiation.

Cadmium, cobalt, hafnium, and the long-lived rare earths are included among the impurities with high neutron absorption. The determination of dysprosium and samarium with a shorter irradiation and that of boron with a technique other than activation analysis is necessary for the complete estimate of the neutron absorption properties of the samples.

An important decrease in the time required for analysis has resulted from setting up the procedure for the radioisotopes in a carrier free state, thus avoiding the effort of making the chemical yield determinations. The recoveries are sufficiently high for practical purposes—i.e., higher than 95%. A further decrease in the operating time has resulted from the automation of the ion exchange separations with a device already described by one of the authors ( $\theta$ ). Seven hours are sufficient to carry out the complete procedure, including the activity measurements. A trained technician may handle more than one analysis at a time. A certain number of elements that give long-lived gamma-emitting isotopes upon neutron activation were not included in the separation scheme. They are generally of minor importance in the analysis of reactor grade aluminum. A semiquantitative estimate of these elements can be made by gamma spectrometry of the separated fractions. The elements will be discussed later.

### EXPERIMENTAL

**Procedure.** The procedure is outlined in Figure 1, and steps one through 15 are described below.

1. Dissolve the irradiated aluminum (approximately 200 mg.) in 3 ml. of 6M HCl and add one drop of concd. HNO<sub>3</sub>. Heat the solution to boiling, then cool in an ice-water bath.

2. Saturate the solution with gaseous hydrogen chloride, then filter with HA Millipore filter. Wash three times with 1-ml. aliquots of 12M HCl.

3. Adsorb the filtrate and washings on an anion exchange resin (Dower 1, X-8, 100- to 200-mesh) equilibrated with 12M HCl. The column is 0.5 cm. in diameter and 10 cm. long (2 ml, of resin). The elution rate must be not higher than 0.2 to 0.3 ml. per minute. Complete the elution of the nonadsorbable metals with 8 ml. of 12M HCl.

4. Elute the groups 1 to 5 with the following solutions: (a) 10 ml. of 12.M HCl-0.5M HF (hafnium and protactinium, traces of cadmium); (b) 10 ml. of 4M HCl (cobalt and neptunium); (c) 20 ml. of 2M HCl-3M HF (iron and tin); (d) 10 ml. of 0.02M HCl (zinc); (e) 10 ml. of water (cadmium). The fractions (b, c, and d) may be analyzed immediately by gamma ray spectrom-



Figure 1. Separation scheme for the determination of long-lived gamma-emitting impurities in reactor grade aluminum

etry. The fractions (a) and (e) are kept for additional processing. The elution rate may be increased to 1 ml. per minute after the elution of the hafnium fraction.

5. Heat the hafnium fraction to boil off the excess of hydrochloric acid. Add two drops of 50% HF and pass the solution on a small Dowex 1 column (0.5 cm. in diameter and 5 cm. long) equilibrated with 6M HCl at a rate of 1 ml. per minute. Wash the column with 3 ml. of 6M HCl, and determine hafnium and protactinium by  $\gamma$ -ray spectrometry of the eluate.

6. Heat the solution of the nonadsorbable metals from step 3 to boil off the excess of hydrochloric acid, then pass the solution on the same column of Dowex 1 at the same rate. Wash the column with 3 ml. of 6M HCl.

7. The column in steps 5 and 6 retains the cadmium that is always present in part in the 12M HCl fractions. This part is particularly high if the elution rate or the loading of the column was too high. Elute the cadmium with 10 ml. of water and count it with the main fraction.

8. Wash the precipitate of  $AlCl_3$  from step 2 with the minimum amount of water to dissolve completely the  $AlCl_3$ .

9. The AlCl<sub>3</sub> solution may contain traces of  $Cr^{+3}$ ,  $Sc^{+3}$ , and rare earths coprecipitated with the AlCl<sub>3</sub>. Add 1 ml. of a 2-mg-per-ml. Fe<sup>+3</sup> solution. Precipitate Fe(OH)<sub>3</sub> with 5*M* NaOH saturated with gaseous chlorine. [Use an excess of the reagent to redissolve the Al(OH)<sub>3</sub>]. Filter on a sintered glass filter and wash the precipitate. Scandium and the rare earths are retained by the iron hydroxide. Chromium is left in the solution as  $Cr^{+6}$ .

10. Dissolve the  $Fe(OH)_3$  on the filter by passing the main fraction of the nonadsorbable metals from step 6. Silver, scandium, the rare earths, the alkali metals, and part of the chromium are present at this point.

11. Add 0.5 gram of AgCl. Heat the solution for a few minutes to complete the exchange between active  $Ag^+$  and inactive AgCl. Filter, wash, and count the filter for  $Ag^{10}$ .

12. Precipitate  $Fe(OH)_3$  with 5M NaOH saturated with gaseous chlorine. Filter on a sintered glass filter and wash the precipitate.

13. Chromium and the alkali metals are present in the alkaline solution from steps 9 and 12. Cool the solutions in an ice-water bath and acidify with the minimum amount of HCl. Pass the solutions on a Dowex 1 column (0.5 cm. in diameter and 5 cm. long) at a rate of 1 ml. per minute. Wash with 3 ml. of water. Cesium and rubidium are determined in the eluate.

14. Take the resin from the column and count the chromium directly on the resin.

15. Dissolve the  $Fe(OH)_3$  from step 12 with 6*M* HCl and analyze the solution for scandium and the rare earths. If further chemical separation between scandium and the individual rare earths is to be applied, remove the iron by passing the solution on a Dowex 1 resin (0.5 cm, in diameter and 5 cm. long) equilibrated with 6M HCl. Wash with 3 ml. of 6M HCl.

### RESULTS AND DISCUSSION

Experimental Approach. A few samples of 99 to 99.9% aluminum were irradiated and analyzed qualitatively. The groups separation by conventional analytical chemistry was applied after addition of carriers. Tentative ion exchange procedures were also applied. A list of possibly present gamma-emitting impurities with long half lives was thus obtained. On the basis of these elements and of the elements of higher importance from a nuclear standpoint, a chemical separation scheme was chosen and tested with radioactive tracers. The scheme was successively modified until the chemical yields were higher than 95% with no interfering activity in other groups.

Adsorbabilities on the Anion Exchange Resin. The adsorbability of the elements that can be separated on the anion exchange resin was measured for the resin batch used in our experiments. The adsorbabilities were determined as distribution coefficients by batch equilibration (12). Again radioactive tracers and gamma counting were used to determine the concentration of the ion. The results are reported in Table II as the ratios of the activity per gram of resin over the activity per milliliter of solution after equilibration. The resin (Dowex 1, X-8, 100- to 200-mesh, Fluka A.G.) was stored dry and weighed at the ambient humidity. The moisture content was determined by heating a sample to constant weight at 100° C.

Irradiations. The irradiations are generally made in the isotope train position of the Ispra I reactor. The thermal flux is approximately  $5 \times 10^{12}$ neutrons per sq.- cm. per second. The standards are known quantities of standardized solutions contained in thin silica vials and irradiated as close as possible to the samples. A neutron flux monitor (a wire of a dilute alloy of cobalt in aluminum) is irradiated with the samples. This permits the correlation of different irradiations and serves as a check on the correct preparation of the standards.

Activity Measurements. The fractions separated with the chemical procedure are analyzed by  $\gamma$ -ray spectrometry. Liquid samples (groups 1, 2, 3, 4, 5, 7, and 8) are contained in small bottles filled at a constant level. The reproducibility of measurements made with different bottles is within 2%. The vials of the standards are broken directly into the counting bottles and diluted to the same level. The silver fraction is counted directly on the filter and compared with a standard prepared by depositing a few drops of the standard solution on a filter paper disk. The chromium group is counted by pouring the resin in the same bottles used for the analysis of liquid samples. The chromium standard is counted in liquid form in the same geometry. The Np<sup>239</sup> in the irradiated uranium standard can be measured without separating the fission products if the irradiation time is of the order of 2 weeks or shorter.

A 3-  $\times$  3-inch NaI(Tl) crystal coupled to a 3-inch DuMont 6363 multiplier phototube was used as the detecting device. The integral line assembly was shielded in a lead cave lined with cadmium and copper as described by Heath (10). The output pulses were analyzed with a 200-channel RIDL pulse height analyzer. A well-type gamma scintillation counter was used for the determination of the adsorbabilities on the anion exchange resin.

Crystallization of the Aluminum Chloride. The crystallization of the aluminum chloride occurs spontaneously in 12M HCl before the anion exchange separation. The adsorption of ions on the surface of the AlCl<sub>3</sub> crystals could be a potential source of error. Fortunately the surface of the crystals when precipitated by gaseous HCl is negatively charged. Adsorption of negatively charged chloride complexes is thus prevented and in practice was found negligible. The anion exchange separations can then be made immediately after two washings.

The trivalent elements that are not

Table II. Distribution Coefficients of the Elements Separated by Anion Exchange

	Eluent						
	12M HCl	12M HCl- 0.5M HF	4M HCl	2M HCl- 3M HF	0.02M HCl	Water	
Hafnium(IV)	186	<1					
Protactinium(V)	710	<1					
Cobalt(II)	56	44	<1				
Neptunium(IV)	$> 10^{3}$	37	<1				
Iron(III)	450	266	<b>54</b>	< 1			
Tin(IV)	>103	$\sim 10^3$	52	7			
Zinc(II)	46	47	890	450	1.5		
Cadmium(II)	10	15	168	87	31	12	

complexed by HCl were adsorbed to some extent. Reprocessing the AlCl<sub>a</sub> as described in the procedure is then necessary before separating chromium, scandium, and the rare earth metals.

Anion Exchange Separations. The distribution coefficients  $(K_d)$  of metals on anion exchange resins are presently known for a wide variety of resins and acids. The knowledge of the adsorbabilities reported in the literature is a great help in planning any separation scheme on resins. Most of them must nevertheless be controlled for the particular batch of resin used. Table II shows the  $K_d$  of the elements separated on the anion exchange resin Dowex 1 with the hydrochloric and hydrofluoric acid concentrations finally chosen in our scheme.

Small columns, containing approximately 1 gram of resin are used in our procedure. The loading of the resin is therefore fairly high when analyzing aluminum with a high impurity content (5 to 8% loading if 0.2 gram of 99% aluminum is analyzed). The use of larger resin columns certainly increases the selectivity, but larger volumes of eluting agent would be required for complete elution. Concentration before measuring the activity would then be necessary, thus increasing the time for analysis.

The adsorption of the metal chloro complexes is made from 12M HCl with a rather low flow rate. The small diffusion coefficients at high ionic strength due to the lower water content of the resin make the equilibration rate low. The increase of the flow rate of the 12M HCl elution would cause a significant spreading of activity along the column and hence a poorer separation. The use of smaller-mesh resins or operation at higher temperature could shorten the time required for the separations at high HCl concentration, but it was not considered necessary.

Hafnium and protactinium are eluted together with 12M HCl-0.5M HF. The adsorbabilities of a number of elements from HCl-HF media were investigated in detail by Nelson, Rush, and Kraus (18). Both elements are completely converted to nonadsorbable fluoride complexes over 4M HCl. Neptunium also is strongly complexed by hydrofluoric acid (22), but the adsorbability with 12M HCl-0.5M HF is sufficiently high to permit a good separation from hafnium and protactinium. Neptunium can be rapidly eluted together with cobalt in the next step (4*M* HCl).

Uranium and thorium are not determined by the activity of the shortlived product of the  $(n, \gamma)$  reaction, but by the activity of the first decay product. The reactions are

```
U^{238} + n \rightarrow U^{239} + \gamma
```

$$U^{239} \xrightarrow{\beta^{-} \operatorname{decay}} \operatorname{Np}^{239}$$

$$\operatorname{Np}^{239} \xrightarrow{\beta^{-} \operatorname{decay}} \operatorname{Pu}^{239}$$

$$\operatorname{Np}^{239} \xrightarrow{\beta^{-} \operatorname{decay}} \operatorname{Pu}^{239}$$

$$\operatorname{Th}^{232} + n \rightarrow \operatorname{Th}^{233} + \gamma$$

$$\operatorname{Th}^{233} \xrightarrow{\beta^{-} \operatorname{decay}} \operatorname{Pa}^{233}$$

$$\operatorname{Pa}^{233} \xrightarrow{\beta^{-} \operatorname{decay}} U^{233}$$

Np<sup>239</sup> and Pa<sup>233</sup> decay with a half life longer than their parents, U<sup>239</sup> and Th233. No equilibrium is therefore attained at any time between parent and daughter activity. The decay curve of the daughter activity also depends upon the saturation of the daughter activity attained during the irradiation in the reactor. Care must be taken when decay corrections are to be applied referring the activity of the sample to the activity of a comparator irradiated simultaneously for quantitative analysis (9). The knowledge of the uranium content of aluminum used for sheathing fuel elements permits the calculation of the amount of fission product activity likely to be found in the moderator or coolant of a nuclear reactor under normal conditions (15).

Iron and tin are both eluted with 2M HCl-3M HF. The distribution coefficients in Table II show that iron must elute before tin. The first 3 ml. of eluate do contain over 95% of the iron and only traces of tin, thus permitting eventually a simple chemical separation within the group.

Zinc and cadmium are eluted with 0.02*M* HCl and water. Cadmium may spread in three fractions: 12*M* HCl  $(K_d = 10)$ , 12*M* HCl-0.5*M* HF  $(K_d = 15)$ , water  $(K_d = 12)$ . Reprocessing of the 12*M* HCl fractions is then necessary. It is sufficient to boil off the excess of HCl to raise the  $K_d$  of cadmium to about 130, thus permitting a rapid separation from the nonadsorbable elements, hafnium and protactinium, on a small Dowex 1 resin column.

Separation within the Nonadsorbable Group. The elements in groups 6 to 9 of Table I are not adsorbed by the anion exchange resin from 12MHCl, and additional chemical separation is necessary for their analysis.

Nelson, Day, and Kraus (17) studied the possibility of using anion exchange resins in chloride form and EDTA as eluting agent. Jervis and Mackintosh (11) used cation exchange chromatography with acidic solutions. Cation resins and complexing elutions could also be used to some extent (4).

Heterogeneous isotopic exchange is used in our procedure to separate silver from the hydrochloric acid solution. The exchange between silver (or halide) ion in solution and a precipitate of silver halide has already been used in analytical radiochemistry (16, 20). The isotopic equilibrium is reached very rapidly and contamination from most ions is negligible.

Nonisotopic carrier precipitation is used in our scheme to separate the rare earths fraction from chromium and the alkali metals. The precipitation of iron hydroxide for quantitative separation of chromium and the rare earths is a well established radiochemical technique (7). Chromium in our procedure is maintained in solution with the alkali metals by oxidation to the hexavalent state. The iron hydroxide can then be immediately processed for the separation of the rare earths if the separation is required. A rapid adsorption-elution cycle on a small Dowex 1 column completely removes the iron carrier. As no carrier has been added for the rare earths a cation exchange-drop elution technique can be used. The separation between the different rare earths can thus be made in about 1 hour (3).

Chromium(VI) is separated from the alkali metals by adsorption on anion exchange resin from slightly acidic solution and is counted directly on the resin. Chromium is adsorbed on the resin as chromate ion. Both elution with sodium hydroxide and with a reducing agent proceeded with difficulty and with excessive tailing. A part of chromium is irreversibly retained on the resin. The same dif-ficulty has been found by others who have tried to elute chromium from cation exchange resins and was attributed to secondary reactions between chromium and the resin structure (19). Direct counting of the resin bed was thus preferred.

Elements Not Included in the Separation Scheme. Table III shows the elements that were not included in the separation scheme, even if they give long-lived gamma-emitting isotopes upon neutron irradiation.

The chemical behavior of most of them in the separation scheme has been tested with radioactive tracers and the results are briefly reported. The gamma spectra of the fractions separated could eventually lead to the semiquantitative estimate of some of them. Their contribution to the longlived gamma activity of neutron-irradiated aluminum is certainly negligible and does not warrant any modification in the separation scheme.

Strontium and barium have a rather small cross section for neutron activation. While the presence of strontium and barium can be detected in aluminum through the shorter-lived isotopes  $Sr^{87}$  and  $Ba^{139}$ , the activity of the longer-lived isotopes  $Sr^{85}$  and  $Ba^{131}$ is negligible. Both elements behave as the alkali metals in the separation scheme and are finally found in group 7. Some barium is lost by coprecipitation with the iron hydroxide in steps 9 and 12.

Zirconium behaves as hafnium and would be found in group 1. The neutron activation cross section of zirconium is 300 times lower than that of hafnium.

The behavior of antimony in the separation scheme largely depends on its oxidation state. Both Sb(III) and Sb(V) are adsorbed on Dowex 1 from 12M hydrochloric acid solution; Sb(III) elutes with 2M HCl-3M HF, while Sb(V) is retained on the resin through the whole procedure and could be analyzed directly on the resin. A quantitative estimate of antimony requires the consideration of the two different geometries (20 ml. of liquid and the resin bed). The error in the determination would be larger than for the isotopes included in the separation scheme. The 0.6-m.e.v.  $\gamma$ -ray of Sb<sup>122</sup> and Sb<sup>124</sup> can be distinguished very well in the gamma spectra of group 3.

The behavior of molybdenum has also been investigated. The distribution coefficients of molybdenum in hydrochloric acid show that Mo(VI) is adsorbed as a negatively charged complex at high molarity. When the molarity is sufficiently low (1 to 2MHCl) to allow the elution of Mo  $(K_d)$ about 10 with 1 to 2M HCl) hydrolysis takes place and molybdenum is retained mechanically by the resin. Elution with 1M HCl gave recoveries from traces to over 80% in a set of experiments. In hydrofluoric-hydrochloric acid mixtures, hydrolysis is prevented; molybdenum has a rather high minimum of adsorption with 2MHCl-1M HF ( $K_d$  about 18) (13). At lower molarity molybdenum is adsorbed as negative fluoride complex. The use of a higher hydrofluoric acid concentration in our separation scheme makes the adsorbability with 2MHCl higher, and in practice molybdenum is retained on the resin through the whole elution procedure, and may be counted on the resin with a chemical yield of 85 to 90%.

A number of metals could be found on the resin with Sb(V) and Mo. Some are retained as strong chloride complexes (ruthenium, palladium, osmium, iridium, platinum, gold, mercury), others as oxy anions (rhenium), and hydrolizable ions (tantalum). The analysis of the resin bed by gamma spectrometry could eventually lead to the determination of some of these In our experiments the elements. activity retained by the resin was generally very low.

Precision. The determination of the chemical yields shows that the error in the chemical separations is generally lower than 5%. Any greater error would be found by contamination of the gamma spectra of the fractions. The error due to neutron flux inhomogeneity is minimized by a

Table III. Long-Lived Gamma-Emitting Isotopes That Could Be Formed upon Neutron Irradiation and Are Not Determined in Separation Scheme Presented

Element	Radio- isotope	Half life	Gamma energy	Chemical behavior in separation scheme
Selenium	Se <sup>75</sup>	127  d	0.08, 0.14, 0.27	
Strontium	Sr85	65 d	0.51	Group 7 (step 13)
Zirconium	Zr95	65 d	0.75	$\frac{1}{3} \frac{1}{3} \frac{1}$
Molybdenum	Mo <sup>99</sup>	67 h	0.14, 0.18, 0.36, 0.74, 0.78	On the resin (step 4)
Ruthenium	Ru <sup>103</sup>	39.8 d	0.50	
Indium	In <sup>114m</sup>	49  d	0.19, 0.55, 0.72	
Antimony	Sb <sup>122</sup>	2.8 d	0.57, 0.68	Sb(III) in group 3 (step 4)
	Sb124	60 <b>d</b>	0.61, 1.68, 2.10	Sb(V) on the resin $(step 4)$
Tellurium	$Te^{123m}$	104 d	0.09, 0.16	
	Te <sup>125m</sup>	58 d	0.03, 0.11	
	$\mathrm{Te}^{129m}$	33.5 d	0.10	
	I131	8.1 d	0.28, 0.36, 0.64	
Barium	Ba131	11.8 d	0.21, 0.24, 0.37, 0.49, 0.62	Group 7 (step 13)
Tantalum	Ta <sup>182</sup>	111 d	0.07, 0.10, 0.15, 0.23, 1.20	On the resin (step 4)
Rhenium	$\mathrm{Re}^{186}$	3.7 d	0.14	
Osmium	O8191	15 d	0.04, 0.13	On the resin (step 4)
Iridium	Ir <sup>192</sup>	74 d	0.31.0.47	On the resin $(step 4)$
Platinum	A 11 199	31d	0 16, 0 21	On the resin (step 4)
Gold	A 11198	2.7 d	0 41	On the regin (step 4)
Manauma	TT~197	2.14	0.00	On the resin (step 4)
mercury	пg Пg-203	4.0 <b>U</b> 47 d	0.00	On the resin (step 4) $On$ the resin (step 4)
	пе	71 (6	0.20	On the result (step 4)

careful packaging of sample and standards before the irradiations, and it was negligible in the irradiation position used. The error due to counting statistics must be of course evaluated for each analysis. The final precision was estimated to be of the order of 8 to 10% relative standard deviation.

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