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December 28, 1950

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PRODUCTION AND ISOLATION OF CARRIER-FREE RADIOISOTOPES*

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December 28, 1950

I. Introduction

In the separation and use of radioactive substances it is frequently necessary to isolate radioisotope preparations which do not contain detectable amounts of stable isotopic material. These preparations, usually designated by the term "carrier-free", are produced principally by nuclear transmutation reactions.[1] Since transmutation products are generally obtained in amounts which are detectable only by their characteristic nuclear radiations, conventional inorganic separation procedures are usually not directly applicable and special methods must be employed. Many of these methods were originally developed in the investigation of the naturally radioactive elements. Even prior to the general acceptance of the theory of isotopy, a great deal of information had already been obtained on the chemical behavior of unweighable amounts of the short-lived radioelements including methods for their separation and isolation. Although few generalizations were then possible, the introduction of the disintegration theory which was soon followed by the arrangement of the radioelements in the periodic table rapidly resulted in a fairly comprehensive

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[1] Non-transmutation nuclear reactions may also be used to produce carrier-free radioisotopes. A successful separation in cases where the target and product are isotopic depends on the removal of the radioactive nucleus from its chemical bond in the target or parent compound as a result of isomeric transition (16,17) or recoil (14,15). These methods although of considerable theoretical interest are at present not generally practical for the production of carrier-free tracers.

understanding of many of the unique chemical, physical and biological properties of unweighable amounts of the radioelements found in the natural decay series.

Probably, however, the most extensive developments in the chemistry of the radioelements in extreme dilution have followed the discovery of artificial radioactivity. The development of the cyclotron and the chain-reacting pile resulted in a tremendous increase in the number and availability of radioisotopes that can be produced in the carrier-free state. And with this, there has been a corresponding increase in our understanding of the fundamental principles of the separation processes. Many new techniques, particularly adapted to carrier-free radioisotope separation, have been developed. These techniques and those previously employed in classical radiochemistry have been successfully used in the separation of carrier-free radioisotopes of most of the stable elements and in the discovery and preliminary chemical identification of several new elements.

The use of carrier-free techniques is essential in certain types of research and highly desirable in many others. It is, of course, fundamentally important to most investigations involving those radioelements, either artificially produced or naturally occurring, of which no stable isotopes have been found in nature. The fact that these substances are normally encountered only in invisible amounts necessarily delimits the type of chemical technique which can be employed in their manipulation. The isolation of carrier-free radioisotopes of the stable elements also is important in several widely differing fields of research. In chemistry they have been used to investigate, (a) the chemical properties of the elements at extreme dilution, (b) adsorption phenomena, and (c) the properties of radiocolloids, in addition to (d) their use as tracers in many chemical problems in which high dilution factors are important. Carrier-free radioisotope preparations are also desirable in certain types of physical measurement; for example, in the characterization of low-energy nuclear radiations, the use of a "mass-less" source is important to insure small self-absorption and in

mass spectrographic determination of nuclear properties to obtain a maximum sensitivity.

The widest use of carrier-free activities and probably one of the most significant is their application to biological and medical research since these preparations may be added to biological systems without changing the mass or concentration of the stable element or compound already present. This situation is extremely desirable, for example, in metabolic studies, in the investigation of trace element functions and in the use of radioisotopes as in vivo radiation sources. An important medical problem involving the metabolism of carrier-free radioisotopes has resulted from the quantity production of plutonium. The need for investigating the radiotoxicological hazards of the fission products and of the transuranium elements, neptunium and plutonium, has necessitated the biological testing of each of the radioisotopes in the carrier-free form in which it would be encountered in the plutonium process. Because of the importance of evaluating the health hazards of these materials a great deal of information has been obtained on the metabolic properties of the radioisotopes produced in fission.

II. Sources of Carrier-Free Radioisotopes

The chain-reacting pile and the cyclotron are the only practical sources of carrier-free radioisotopes although a few useful radioisotopes are still obtained from the natural decay series. The important nuclear reactions for each of these modes of production are described briefly below.

Pile Reactions: Neutrons at pile energies produce radioisotopes in the carrier-free state by four main types of nuclear transmutation reactions.

(1) Nuclear fission: In the comprehensive investigation of the radioactive species formed in the fission of uranium more than 160 radioactive isotopes have been identified ranging in the case of U^{235} fission from Z = 30 (zinc) to Z = 63 (europium), and from A = 72 to A = 158. As a result of the fact that the neutron excess of U^{235} is considerably greater than that required for stability in the fission-product region, the primary fission-product nuclei achieve stability through

successive β^- decay giving rise to fission product chains. The radioisotopes in each chain, their relationships, their mass assignment and yields have been determined in most cases (1,2). Although these radioisotopes are by-products of normal pile operation, only a few have half-lives sufficiently long to warrant their routine separation. The major radioactive fission products of half-life greater than one week are described in Table I. Most of these are available in carrier-free form from the Isotopes Division, Atomic Energy Commission, Oak Ridge, Tennessee (3).

(2) Neutron capture (n,γ) followed by β emission: With the exception of the (n,f) reactions, the (n,γ) reaction is the most important source of carrier-free radioisotopes in the pile. Although the (n,γ) reaction alone results in the production of radioisotopes which are isotopic with the target element and hence not separable in the carrier-free state, it may be used to produce carrier-free radioisotopes if the product isotope decays to give a radioactive descendant.

Table II describes the carrier-free radioisotopes which are routinely produced in the pile using this type of nuclear reaction. The activation cross section of the target isotope and the characteristics of the radioactive descendant are also listed.

(3)(4) Neutron capture with proton and alpha particle emission, $(n,p)(n,\alpha)$ reaction: A few important (n,p) and (n,α) transmutation reactions are possible with pile neutrons. These are confined to the production of radioisotopes of the lighter elements because only a negligible fraction of pile neutrons have energies sufficient to overcome the potential barrier to change particle emission above Z = 20. Resonances in certain nuclei also permit (n,p) and (n,α) reactions to proceed with neutrons of low energy. Table II lists the radioisotopes which are produced by (n,p) and (n,α) reaction in the pile.

TABLE I

MAJOR RADIOACTIVE FISSION PRODUCTS OF HALF-LIFE GREATER THAN 1 WEEK (1,2,3)

Radioisotope	Half-Life	Energy (Mev) Beta	Gamma	Relative Yield (Curies)*
Sr^{89}	55 d	1.48	none	1.0
Sr^{90}	~30 y	0.65	none	0.035
Y^{91}	57 d	1.53	none	1.25
Zr^{95}	65 d	0.394(98%)	0.73(93%)	1.35
Cb^{95}	35 d	0.154	0.75	1.7
Tc^{99}	$\sim 5 \times 10^{5}$ y	0.32	none	0.7×10^{-6}
Ru^{103}	42 d	0.3 (95%)	0.56	0.9
Ru^{106}	1.0y	~0.03	none	0.065
Te^{127m}	90 d	I.T.	0.086	0.0075
Te^{129m}	32 d	I.T.	0.102	0.03
I^{131}	8 d	0.35;0.60	0.63;0.36;0.28	0.065
Cs^{137}	33 y	0.55;0.84	0.66	0.032
Ba^{140}	12.5d	1.05	0.529	0.35
Ce^{141}	28 d	0.66	0.2	1.0
Ce^{144}	275 d	0.3	none	0.85
Pr^{143}	13.8d	0.83	none	0.4
Nd^{147}	11 d	0.90	0.55	0.12
Pm^{147}	3.7y	0.22	none	0.14

*Relative amounts of fission products co-existing one month from the end of a several-month exposure. Ref. (1)

TABLE II

A- CARRIER-FREE RADIOISOTOPES PRODUCED IN THE PILE BY (n,γ) REACTION FOLLOWED BY DECAY (2,3,4)

Radioisotope	Half-Life	Target Isotope	Percent Abundance	Thermal Neutron Activation Cross Section (Barns) Isotope	Natural	Half-Life of (n,γ) Product
33 As^{77}	40 h	Ge^{76}	6.5	0.085	0.0055	12 h
41 Cb^{95}	35 d	Zr^{94}	17.4			65 d
43 Tc^{97m}	93 d	Ru^{96}	5.68		0.15	2.8d
43 Tc^{99}	9.4×10^5 y	Mo^{98}	24.1	0.415	0.10	67 h
45 Rh^{105}	37 h	Ru^{104}	18.27		0.122	4.4h
47 Ag^{111}	7.5d	Pd^{110}	13.5	0.39	0.0525	26 m
51 Sb^{125}	2.7y	Sn^{124}	6.8	0.574	0.039	9 m
53 I^{131}	8.0d	Te^{130}	34.52	0.222	0.0735	25 m
55 Cs^{131}	10.2d	Ba^{130}	0.101			12.0d
59 Pr^{143}	13.9d	Ce^{142}	11.7			33 h
61 Pm^{147}	3.7y	Nd^{146}	16.5			11.0d
61 Pm^{149}	47 h	Nd^{148}	6.8			1.7h
63 Eu^{155}	2 y	Sm^{154}	22.53		1.10	25 m
79 Au^{199}	3.3d	Pt^{198}	7.2	3.92	0.292	31 m
84 Po^{210}	140 d	Bi^{209}	100	0.015	0.015	5.0d

B- CARRIER-FREE RADIOISOTOPES PRODUCED IN THE PILE BY (n,p) AND (n,α) REACTION

Radioisotope	Half-life	Produced by	Reference
H^3	12 y	Li(n,α)	(55,57)
C^{14}	5100 y	N(n,p)	(64,65,66)
P^{32}	14.3d	S(n,p)	(78,79,80,81,82)
S^{35}	87.1d	Cl(n,p)	(83,84,79,67)
Ca^{45}	180 d	Sc(n,p)	(87,88)

Cyclotron Reactions

The cyclotron is the only source of a large number of useful carrier-free radioisotopes which are not available from pile sources (2,3). In addition, although a desired radioisotope may be produced in the pile, half-life and other technical limitations may make cyclotron irradiation the more practical method of production.

With the exception of the (n,f) reaction, charged particle bombardments differ from pile reactions in the greater probability of concurrent reactions. This is particularly true at the higher bombardment energies. With 20 Mev deuterons, for example, three and occasionally four radioactive elements may be produced concurrently from the target element (8). At energies considerably higher than this ($\sim$ 100 Mev) a whole spectrum of transmutation products are observed.

Reaction products covering a range from the region of the target nucleus down to nuclei about 20 mass units lighter are produced by bombardment with particles having energies in the hundred million volt range (9,10,140). To indicate these reactions in which excited nuclei are degraded by losing one or more nucleons, the term spallation has been suggested. The majority of the artificially produced tracers are produced by the relatively simple reactions involving the emission of not more than three or four nuclear particles.

Because of the great variety of nuclear transmutation reactions (2,5) which can be brought about by deuteron, proton and alpha particle bombardment, it is frequently possible to produce a desired radioisotope by several different types of nuclear reactions. In determining the particular reaction to be employed, several factors must be considered, including (1) the yield of the desired radioisotope, (2) the relative yields of the possible concurrent side reactions, (3) the problem of the ultimate chemical separations of the desired product, and (4) the chemical and physical limitations which must necessarily be imposed in the choice of target material for charged particle bombardment.

Considerations (1) and (2) are discussed below and (3) and (4) are covered in the section on separation procedures.

Deuteron Reactions

For the production of many useful radioisotopes in the carrier-free state the reaction types (d,α)(d,2p)(d,n)(d,2n) are the most important. The (d,α) and (d,2p) reactions are most useful in producing radioisotopes of lower atomic number because of the potential barrier to charged particle emission which increases with atomic number. Although the yield of these reactions increases with deuteron energy, at the higher energies the (d,n) and (d,2n) reactions become relatively more important for the elements of higher atomic number. The (d,αp) reaction is relatively unimportant in the production of practical amounts of radioisotopes although the possibility of this reaction must be considered as a source of radioactive contamination.

In practice, the above transmutation reactions and the non-transmutative (d,p) reaction all occur concurrently with reaction probabilities determined by the energy of the bombarding deuteron and by the atomic number of the target nuclei. The reaction probability (σ) of competing reaction types and their energy dependence (thin-target yields) have been determined for a number of target elements using the stacked-foil technique (30). Nuclear reactions for which excitation functions (σ versus E) have been determined are listed with references in Table III. In radioisotope production by charged particle bombardment the target is generally thick enough to absorb the entire beam. Thick target yields may be obtained by graphical integration of the particular excitation function involved (30), although usually they are obtained directly from thick-target bombardments. Available thick-target yields for charged particle reactions are given in Table IV.

Alpha Particle (Helium ion) Reactions

Alpha-particle capture followed by neutron emission, (α,n)(α,2n)(α,3n) reaction, is useful in the production of certain radioisotopes, particularly of the heavier elements. The principal advantage of this type of reaction is the double increment in atomic number. The (α,xn) reaction is important, for example,

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in the preparation of radioisotopes of astatine (Z = 85) because the only practical target element for this reaction is bismuth (Z = 83). The reactions of the type (α,p) and (α,pn) are more probable with elements having low atomic numbers because of the potential barrier to charged particle emission. Excitation functions for alpha-particle reactions are included in Table III. Thick target yields are given in Table IV.

Proton Reactions

All of the transmutation reactions induced by proton bombardment can be duplicated, as far as carrier-free radioisotope production is concerned, by deuteron bombardment. Deuterons are usually favored over protons because of general operation considerations and because higher energies are more readily obtained. Yield data for a few proton reactions are given in Tables III - IV.

TABLE III

EXCITATION FUNCTIONS OF TRANSMUTATION REACTIONS

TABLE OF REFERENCES

Target	Reference	Transmutation Reactions	Max. Energy (Mev)
C	(33)	$C(d,n)N^{13}$	5
N	(43)	$N(n,p)C^{14}$	
	(53)	$N(n,p)C^{14}$; $N(n,\alpha)B^{11}$	1.7
O	(26)	$O(p,n)F^{18}$	4
Mg	(31)	$Mg(d,\alpha)Na^{24}$	3.5
	(37)	$Mg(d,\alpha)Na^{22,24}$	14
Al	(45)	$Al(d,\alpha p)Na^{24}$	14
S	(48)	$S(n,p)P^{32}$	5.8
Cl	(42)	$Cl(n,p)S^{35}$	
	(42)	$Cl(d,\alpha)S^{35}$	14
Fe	(29)	$Fe(d,n)Co^{55}$	10

TABLE III, CONT'D

Target	Reference	Transmutation Reactions	Max. Energy (Mev)
Fe	(40)	$Fe(d,n)Co^{55}$; $Fe(d,\alpha)Mn^{52}$	10
Ni	(24)	$Ni(p,n)Cu^{61,62,64}$	6.3
	(28)	$Ni(p,n)Cu^{64}$	4
	(32)	$Ni(d,n)Cu^{61}$	5
Cu	(24)	$Cu(p,n)Zn^{63}$	6.3
	(27)	$Cu(d,2n)Zn^{63}$	16
	(28)	$Cu(p,n)Zn^{63}$	4
	(52)	$Cu(d,2n)Zn^{63,65}$;$Cu(d,\alpha)Ni^{63}$	14
Zn	(25)	$Zn(p,n)Ga^{64,68,70}$	6.3
	(26)	$Zn(p,n)Ga^{68,70}$	4
Se	(25)	$Se(p,n)Br^{80,82}$	6.3
	(36)	$Se(p,n)Br^{80,82}$	4.0
Br	(30)	$Br(d,2n)Kr^{79}$	13.5
Rh	(47)	$Rh(\alpha,n)Ag^{106}$;$Rh(\alpha,2n)Ag^{105}$	19
Pd	(34)	$Pd(d,n)Ag^{106}$; $Pd(d,2n)Ag^{106}$	6
Ag	(36)	$Ag(d,2n)Cd^{107,109}$	9
	(47)	$Ag(\alpha,n)In^{112}$;$Ag(\alpha,2n)In^{111}$	19
	(49)	$Ag(\alpha,xn)In^{109,110,111}$	37
In	(51)	$In(\alpha,n)Sb^{118}$;$In(\alpha,2n)Sb^{117}$;$In(\alpha,3n)Sb^{116}$	38
Au	(37)	$Au(d,2n)Hg^{197}$	9
Tl	(39)	$Tl(d,n)Pb^{206}$	9
Pb	(38)	$Pb(d,n)Bi^{207}$	9
Bi	(35)	$Bi(d,n)Po^{210}$	9
	(39)	$Bi(d,n)Po^{210}$	9
	(50)	$Bi(d,n)Po^{210}$;$Bi(d,3n)Po^{208}$	19
	(46)	$Bi(d,n)Po^{210}$	14.5
	(51)	$Bi(\alpha,2n)At^{211}$;$Bi(\alpha,3n)At^{210}$	38

TABLE IV

A- THICK TARGET YIELDS OF SOME ARTIFICIAL RADIOELEMENTS -Ref. (70)

Yields are given in terms of microcuries per microampere hours. The term microcurie refers to the absolute number of disintegrations per second of the artificially prepared radioelement: 3.7×10^4 disintegrations per second. This definition, however, does not apply to those radioelements whose yield values are enclosed in parenthesis since they are all substances decaying by K capture and the measured radiation contains varying proportions of x-rays, gamma rays, and internally converted electrons. In such instances, the value of the microcurie is a comparable one and simply indicates that the amount of ionization produced is equivalent with the measuring device employed to that from one microcurie of the radioactive standard. A Lauritsen electroscope with a thin-walled aluminum window was employed for these measurements. The total air equivalent of the window together with the distance of the sample from the instrument totalled 4 cm. The instrument was calibrated by means of UX_1 standards which were covered with a sufficient thickness of aluminum foil to screen out most of the soft UX_1, betas and approximately 15% of the more energetic UX_2 beta particles. The values presented in the table include yields for deuterons at energies of 8, 14, and 19 Mev, and helium ions at 38 and 44 Mev. The last column gives the factor of the differences of isotopic abundances of the target materials. This correction was not applied to the yield data. In the case of C^{14}, the value given is a calculated one assuming a bombardment of 100 gallons of saturated solution of ammonium nitrate. The accuracy of the underlined yield values is believed to be accurate to plus or minus 25%. Those not underlined are subject to much greater error.

TABLE IV, CONT'D

Isotope	Half-Life	Radiations	Reaction	Yield in μc per μa Hr. 8 Mev D	14 Mev D	19 Mev D	38 Mev α	44 Mev α	F
1 H^3	31 y	β^-	Be-d-H^3	---	0.1	---	---	---	1
4 Be^7	52.9d	K,γ	Li-d-2n	---	---	2			
6 C^{11}	21 m	β^+	B-d-n	500	---	---	---	---	5
6 C^{14}	25,000 y	β^-	N-n-p	---	.0005*	---	---	---	1
7 N^{13}	9.93m	β^+,γ	C-d-n	1000	---	---	---	---	1
11 Na^{22}	3 y	β^+,γ	Mg-d-α	---	1.0	---	---	---	9
11 Na^{24}	14.8 h	β^-,γ	Na-d-p	---	---	1500	---	---	1
12 Mg^{27}	10.2 m	β^-,γ	Mg-d-p	500	2800	---	---	---	9
15 P^{32}	14.3 d	β^-	P-d-p	30	120	---	---	---	1
16 S^{35}	88 d	β^-	Cl-d-α	---	---	8.4	---	---	4
17 Cl^{38}	37.0 m	β^-,γ	Cl-d-p	1000	---	---	---	---	4
19 K^{42}	12.4 h	β^-,γ	K-d-p	---	50.0	107	---	---	15
19 K^{42}	12.4 h	β^-,γ	A-α-pn	---	---	---	---	80	1
19 K^{42}	12.4 h	β^-,γ	Ca-d-α	---	---	3.3	---	---	50
20 Ca^{45}	180 d	β^-	Ca-d-p	---	0.01	0.05	---	---	50
25 Mn^{54}	310 d	K,γ	Fe-d-α	---	(1.0)	---	---	---	1.1
26 Fe^{59}	47 d	β^-,γ	Fe-d-p	---	0.03	---	---	---	360
26 Fe^{55}	4 y	K,e^-.	Mn-d-2n	---	---	(0.02)	---	---	17
27 Co^{57}	270 d	K,e^-,γ	Fe-d-n	---	(1.0)	(5.0)	---	---	1.1
29 Cu^{64}	12.8 h	β^-,β^+	Cu-d-p	---	3000	---	---	---	1.5
30 Zn^{65}	250 d	β^+,K,e^-,γ	Cu-d-2n	---	(0.5)	---	---	---	3
33 As^{74}	16 d	β^+,β^-,γ	Ge-d-n	---	2	10	---	---	3
37 Rb^{86}	19.5 d	β^-,γ	Rb-d-p	---	4.3	---	---	---	1.4

*Calculated from known neutron yields and ρ_a for N in a saturated solution of NH_4NO_3 of 100 gallons.

TABLE IV, CONT'D

Isotope	Half-Life	Radiations	Reaction	Yield in μc per μa Hr. 8 Mev D	14 Mev D	19 Mev D	38 Mev α	44 Mev α	F
37 Rb^{86}	19.5 d	β^-,γ	Sn-d-α	---	1.0	---	---	---	1.2
38 Sr^{85}	66.5 d	K,e^-,γ	Rb-d-2n	---	(0.13)	(0.60)	---	---	1.2
38 Sr^{89}	55 d	β^-	Sr-d-p	---	10.4	---	---	---	1.2
39 Y^{86}	108 d	K,e^-,γ	Sr-d-2n	---	(0.10)	(1.0)	---	---	10
40 Zr^{89}	81 h	β^+	Y-d-2n	---	7.0	75.0	---	---	1
40 Zr^{95}	63 d	β^-,γ	Zr-d-p	---	.15	---	---	---	6
41 Cb^{90}	18 h	β^+	Mo-d-α	---	2.4	---	---	---	6
41 Cb^{95}	36 d	β^-,γ	Mo-d-α	---	.05	---	---	---	11
41 Cb^{91}	60 d	K,e^-,γ	Zr-d-n	---	(1.0)	---	---	---	2
52 Te^{121}	125 d	K,e^-,γ	Sb-d-2n	---	(0.16)	---	---	---	1.8
53 I^{131}	8 d	β^-,γ	Te-d-n	1.0	20.0	---	---	---	3
56 Ba^{133}	39.6 h	K,e^-,γ	Cs-d-2n	---	(100.0)	---	---	---	1
84 Po^{210}	136 d	α	Bi-d-n	---	2.0	---	---	---	1
84 Po^{210}	136 d	α	Pb-α-2n	---	---	---	---	0.85	2
85 At^{211}	7.5 h	α	Bi-α-2n	---	---	---	1.0	1000	1
91 Pa^{233}	27.4 d	β^-,e^-,γ	Th-d-2n	---	- -	(20.8)	---	---	1

TABLE IV

B- THICK TARGET YIELDS FOR SOME NUCLEAR REACTIONS OBTAINED WITH 14 MEV DEUTERONS (Ref. 22)

I ISOTOPE	II HALF-LIFE	III RADIATION	IV REACTION	V rd/μah YIELD*
11 Na^{22}	3.0 yr.	β^+, γ	$Mg^{24}(d,\alpha)$	0.065
11 Na^{24}	14.8 hr.	β^-, γ	$Na^{23}(d,p)$	410
11 Na^{24}	14.8 hr.	β^-, γ	$Mg^{26}(d,\alpha)$	8.70
11 Na^{24}	14.8 hr.	β^-, γ	$Al^{27}(d,p\alpha)$	1.66
14 Si^{31}	170 min.	β^-	$Si^{30}(d,p)$	37
15 P^{32}	14.3 day	β^-	$P^{31}(d,p)$	8.5
17 Cl^{38}	37 min.	β^-, γ	$Cl^{37}(d,p)$	1500
19 K^{42}	12.4 hr.	β^-	$K^{41}(d,p)$	8.3
25 Mn^{52}	6.5 day	β^+, K, γ	$Cr^{52}(d,2n)$	3
27 Co^{60}	5.3 yr.	β^-, γ	$Co^{59}(d,p)$	0.040
29 Cu^{64}	12.8 hr.	β^-, β^+, K	$Cu^{63}(d,p)$	92
30 Zn^{63}	38 min	β^+	$Cu^{63}(d,2n)$	1160
30 Zn^{65}	250 day	K, γ	$Cu^{65}(d,2n)$	0.126
35 Br^{82}	34 hr.	β^-, γ	$Br^{81}(d,p)$	28.5
36 Kr	34 hr.	β^+, γ	Br(d,2n)	190μgRa
38 Sr^{89}	55 day	β^-	$Sr^{88}(d,p)$	1.3
39 Y^{88}	87 day	K, γ	$Sr^{88}(d,2n)$	1.4
53 I^{130}	12.6 hr.	β^-, γ	$Te^{130}(d,2n)$	32
53 I^{131}	8.0 day	β^-, γ	$Te^{130}(d,n)$	3.2

*1 rd = 10^6 disintegrating atoms per second.

III. General Considerations in Carrier-Free Radioisotope Preparation. Types of Separation Processes

Although the detailed methods which have been developed for isolating carrier-free radioelements are extremely varied and depend on the particular problem at hand, all are based on a relatively few general types of separation processes; (a) co-precipitation, (b) leaching, (c) radiocolloid formation, (d) electrodeposition, (e) ion-exchange, (f) solvent extraction, (g) distillation (volatilization). Each of these is discussed briefly below in terms of its applicability to the problem of isolating unweighable amounts of the probe carrier-free radioelement, or compound of the element, in aqueous solution at a pH range of 3 to 8. This criterion has been adopted for two reasons. First, because the most extensive use of carrier-free radioisotope preparation is in biological and medical usage for which it is usually desirable to obtain radioactivities of extremely high chemical, radiochemical and isotopic purity in isotonic solution of sodium chloride, sodium sulfate or other non-toxic salt. Second, because these specifications are generally high enough to meet requirements of almost all other types of investigation requiring carrier-free radioisotopes.

(a) Co-precipitation: The separation of sub-microgram quantities of radioelements from solution by means of non-isotopic precipitates usually involves a subsequent separation of the radioelement from the carrier precipitate. It is desirable, therefore, to use as co-precipitating agents, substances which can be easily separated with a minimum amount of chemical and physical manipulation. True co-precipitation processes (6,7), (i.e., those cases in which the radioelement because of similar chemical properties is incorporated in the precipitate through isomorphism or mixed crystal formation) although to a large extent extremely reproducible and independent of precipitating conditions are not of great practical importance particularly in the isolation of radioisotopes for biological research. The inherent difficulties involved in the subsequent separation of the chemically

similar radioelement and precipitant frequently more than offset the desirable features of co-precipitation processes. This factor has, however, become somewhat less limiting with the development of the ion-exchange technique (108).

More generally in the separation of carrier-free activities a "scavenging" type of precipitation reaction is employed. In these cases, the radioelement is carried down as a result of adsorption phenomena. Although precipitation reactions of this type are quite sensitive to changes in experimental conditions and are not as specific as two co-precipitation reaction, they have the important advantage in that the scavenging precipitate can usually be chosen so that its subsequent separation from the carrier-free radioisotope involves a minimum amount of manipulation. Table V is a resume of some important co-precipitation - scavenging types of precipitation reactions. A serious disadvantage of co-precipitation processes in general is the fact that they involve the addition of macro amounts of carrier material which may contain impurities which are isotopic with the desired radioisotope. In addition, co-precipitation reactions, particularly those involving adsorption phenomena, are frequently specific only under rigidly controlled experimental conditions (7).

(b) Leaching: The extraction of carrier-free radioisotopes into aqueous solution from macro quantities of insoluble target or parent material has been used as a separation procedure in a few cases. These are described in Table V . Although quantitative separations are rarely obtained, the relative simplicity of the leaching technique makes it a useful practical procedure. Since the carrier-free radioelement must exist, under the particular experimental conditions, as a soluble compound and must not be preferentially adsorbed or incorporated in the insoluble material, the separation has had only a limited application.

(c) Radiocolloid Formation: Carrier-free radioelements under conditions which normally result in the formation of visible precipitates if a sufficient quantity of the material is present, may form radiocolloidal aggregates even though the solubility product conditions are not satisfied. Although the exact nature of

this phenomena is not entirely clear, it has been suggested (6,58) that the radioelement becomes adsorbed on colloidal impurities which are normally present in solution. Adsorption may also occur on the walls of the containing vessel. This phenomena has been satisfactorily employed in the isolation of carrier-free radioisotopes of several elements, particularly those forming sparingly soluble hydroxides; an alkaline "solution" of the carrier-free activity is passed through filter paper or sintered glass which removes the radiocolloidal material by preferential adsorption. The invisible quantity of adsorbed radioelement is washed with water and then removed with dilute acid. Radioisotopes which have been separated using this technique are shown in Table V. The procedure is particularly useful in the isolation of radioactive tracers for biological investigation since an isotonic saline solution may be obtained simply by neutralizing the hydrochloric acid wash.

(d) Electrodeposition: Radioelements which have been isolated in the carrier-free state by methods involving electrodeposition reactions include, (1) electronegative elements which are reduced to the metallic state by displacement with a more electropositive element or by an applied electromotive force, (2) elements which form insoluble oxides by anodic oxidation reactions, (3) elements which form insoluble compounds with the electrode material as a result of either cathodic or anodic reactions. Carrier-free radioisotopes which have been separated by electrodeposition reactions are given in Table V. Separation of the activity from the electrode material, particularly when platinum or other noble metals are used, is usually accomplished by preferential dissolution of the radioelement. With a mercury cathode, the separation is conveniently accomplished by volatilization. In certain cases, however, the deposited activity can be removed only by treatment with chemical reagents which react with theelectrode material necessitating a subsequent separation to obtain a chemically pure solution of the activity suitable for use in biological systems.

(e) Ion exchange: The separation of trace amounts of radioelements by selective elution from ion exchange columns has become an exceedingly important process in the preparation of carrier-free radioisotopes, particularly in the case of the cationic fission product elements. Although ion-exchange methods had been previously used, the separations obtained were not sufficiently good to warrant the use of exchange adsorption phenomena in radioisotope separations until the important effect of complexing agents on the adsorption-elution cycle was recognized. The necessity of isolating pure fission products for biological testing resulted in the development of this entirely new technique of ion-exchange separation which has had its most important application in the separation of carrier-free radioisotopes of the rare earth elements from fission product mixtures (108). In this work it was found that a mixture of carrier-free fission product activities adsorbed onto amberlite or dowex 50 ion-exchange resin could be selectively eluted with a dilute aqueous solution of organic complexing acids buffered to a controlled pH with ammonium hydroxide. In a typical separation, a dilute hydrochloric acid solution of carrier-free fission products is passed through a column of Amberlite IR-1 or Dowex 50 ion-exchange resin which adsorbs the activities in a narrow band at the top of the column. After washing thoroughly with water, the carrier-free radioisotopes are selectively eluted with 5 percent citric acid solution, at a pH of from 3 to 8 depending upon the type of resin, the rate of removal and the separation desired. Under optimum conditions, the method is sufficiently precise so that a quantitative separation of neighboring rare earth elements may be obtained. To obtain the carrier-free radioisotopes in hydrochloric acid solution, the citric acid effluent is acidified to reduce the complexing action of the citrate ion and passed through a second column which readsorbs the activity. After washing with dilute hydrochloric acid, the activity is stripped from the column with 6 normal hydrochloric acid.

The ion-exchange technique is the only practical method available for separating many of the fission produced radioisotopes in high purity and in a form which may be used in biological investigation with a minimum amount of chemical

treatment and manipulation. The important ion-exchange separations are summarized in Table V.

(f) Solvent Extraction: The selective extraction of a radioactive tracer as a non-polar compound or complex from an aqueous solution by an immissible organic solvent is frequently the most satisfactory method of separating the activity from non-isotopic substances which may be present in either macro or micro concentrations. These processes can often be made highly selective. Solvent extraction is also used to remove macro quantities of non-isotopic substances from an aqueous solution of a desired carrier-free radioelement. This method of removing a macro constituent from solution is of great importance when separation by precipitation would result in a substantial loss of the carrier-free radioelement by co-precipitation.

As a rule, the distribution ratio of an extractable substance is more or less independent of the initial concentration, but the fact that a favorable distribution ratio is obtained at macro or even micro concentration levels does not necessarily mean that sub-micro amounts of the substance will be extractable. For example, carrier-free radio-iron cannot be extracted into ethyl ether from 6N hydrochloric although, as is well known, the distribution ratio of ferric chloride at higher concentrations is sufficiently large to permit the use of ether extraction as a quantitative separation process. This concentration effect has been observed in other cases also and is generally assumed to indicate that the molecular weight of partitioning substance is greater in the organic solvent as a result of polymerization. Usually, however, carrier-free radioelements at concentrations as low as 10^{-15} M show extraction coefficients which are of the same order of magnitude as those obtained at the macro level.

A number of organic substances have been used to form extractable non-polar complexes or chelates with carrier-free radioelements. Compounds which form soluble non-polar complexes whose dissociation constants or pH dependent are particularly useful in separating a carrier-free radioelement from a mixture. Extraction separations are summarized in Table V.

(g) Distillation (volatilization): Carrier-free radioelements which have been separated from solutions, melts and solids by volatilization or distillation processes are shown in Table V . It has been found that the experimental conditions for the volatilization of a carrier-free radioelement from solutions and melts, i.e., from a homogeneous liquid phase, are roughly equivalent to those found to be optimum for macro quantities of that element, although an inert carrier-gas is generally required. In volatilization separation from solids, the relative behavior of sub-macrogram amounts of a radioelement is to a great extent dependent on the nature of the solid and its physical state. In some cases, carrier-free radioisotopes of the inert gases produced by transmutation reactions in solids can be volatilized only after the solid target or parent material has been fused or dissolved in aqueous solution (85,105).

Under proper experimental conditions, very sharp separations can often be obtained by volatilization methods. These procedures are more generally applicable, however, to the separation of carrier-free activities from solutions and melts than from solids. An important advantage, particularly in the isolation of radioisotopes for biological research, is that the activity can usually be condensed in a small volume of water or aqueous solution which can be used with a minimum amount of additional chemical treatment.

TABLE V

CLASSIFICATION OF SEPARATION PROCESSES USED IN THE ISOLATION OF CARRIER-FREE RADIOISOTOPES
REFERENCES TO THE DETAILED METHODS OUTLINED IN TABLE VI ARE GIVEN IN PARENTHESES

Radioisotope	Produced by	Separation Processes which Have been Employed: Co-precipitation	Solvent Extraction	Ion Exchange	Gas Evolution Vaporization Distillation	Radiocolloid	Leaching	Electro-deposition
H^{3}	Be(d,2α) Li(n,α)				(55,56,57)			
Be^{7}	Li(d,n)	(58,59)	(58,59,61)			(60)		
C^{11}	B(d,n)				(62,63)			
C^{14}	N(n,p)				(64,65,66)			
N^{13}	C(d,n)				(68,69)			
F^{18}	O(α,pn)						(70)	
$Na^{22,24}$	Mg(d,α)			(76)			(72,73,74,75)	
Mg^{27}	Al(d,2p)					(77)		
P^{32}	S(n,p)	(78,79)	(80,81)	(78)				(82)
S^{35}	Cl(d,α) Cl(n,p)			(67)			(83,84,79)	
A^{37}	Cl(d,2n)				(85)			
$K^{42,43}$	A(α,pn)						(86)	
Ca^{45}	Sc(n,p) Sc(d,2p)	(87)	(88)				(87)	
Sc^{46}	Ti(d,α)					(89)		
V^{48}	Ti(d,2n)						(90)	
Cr^{51}	V(d,2n)	(91)						
$Mn^{52,54}$	Fe(d,α)	(92)	(92)					
$Mn^{52,54}$	Cr(d,n)	(93)	(93)					

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Radioisotope	Produced by	Co-precipitation	Solvent Extraction	Ion Exchange	Gas Evolution Vaporization Distillation	Radiocolloid	Leaching	Electro-deposition
Fe^{59}	Co(d,2p) Co(n,p)		(94)					
$Co^{56,57,58}$	Fe(d,xn)						(92)	
$Cu^{64,67}$	Zn(d,2p) Zn(n,p)	(97)	(95,96)					(98)
Zn^{65}	Cu(d,2n)		(99)				(99)	
Ga^{67}	Zn(d,xn)		(100)					
Ge^{71}	Ga(d,2n)							
As^{74}	Ge(d,2n)				(101,102)			
Se^{75}	As(d,2n)	(103)			(103)			
$Br^{80,82}$	Se(d,2n)				(104)			
Kr^{79}	Br(d,2n)				(105,106)			
Sr^{85}	Rb(d,2n)	(107)	(107)					
$Sr^{89,90}$	U(n,f)			(108,109)				
Y^{88}	Sr(d,2n)					(110)		
Y^{91}	U(n,f)			(108,111)				
Zr^{89}	Y(d,2n)	(107)						
Zr^{95}	U(n,f)	(112,107)	(112)	(108)				
Cb^{95}	U(n,f)	(112,107)	(112)					
Mo^{99}	Zr(α,n)		(113)					
Tc^{95}	Mo(d,n)				(114,115)			
Tc^{99}	U(n,f)	(116)			(116)			
Ru^{103}	U(n,f)	(107)			(107)			

Radioisotope	Produced by	Co-precipitation	Solvent Extraction	Ion Exchange	Gas Evolution Vaporization Distillation	Radiocolloid	Leaching	Electro-deposition
Pd^{103}	Rh(d,2n)	(117)			(117)			
$Ag^{105,106,111}$	Pd(d,2n)	(118)						
Ag^{111}	Pd^{111} β decay							(119)
Cd^{109}	Ag(d,2n)	(120)						
$In^{111,114}$	Cd(α,pn)	(121)						
Sn^{113}	Cd(α,xn)				(113)			
$Sb^{120,122}$	Sn(d,xn)	(122)			(122)			
$Te^{127,129}$	U(n,f)	(107)			(107)			
I^{131}	Te(d,xn)	(107)			(123,107)			
I^{131}	U(n,f)				(124,67)			
I^{131}	Te β decay				(125,124,67)			
X^{127}	I(d,2n)				(107,85)			
Cs^{131}	Ba^{131} β decay						(127)	
Cs^{137}	U(n,f)	(128,107)		(127)				
Ba^{133}	Ca(d,2n)						(129,107)	
Ba^{140}	U(n,f)			(108,130)				
La^{140}	Ba^{140} β decay	(107)	(107)	(108,130)				
Rare Earths Z=58-Z=63	U(n,f)			(108,127,67)				

Radioisotope	Produced by	Co-precipitation	Solvent Extraction	Ion Exchange	Gas Evolution Vaporization Distillation	Radiocolloid	Leaching	Electro-deposition
Rare Earths Z=64-Z=71	Z(p,xn) (d,xn) (α,xn)	(131)		(131)				
$Ta^{177,178,180}$	Hf(d,xn)	(132)		(132)				
$Re^{183,184}$	Ta(α,xn)	(133,134)			(133,134)			
Os^{185}	W(α,xn)		(135)					
$Bi^{204,206}$	Pb(d,2n)					(136)		
Po^{210}	Pb(α,xn)	(137)	(137)					
Po^{210}	Bi(d,n)	(137)	(137)					
$At^{210,211}$	Bi(α,xn)				(138,139)			

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Purity Considerations

Target Materials and Reagents: In preparing carrier-free radioisotopes of the stable elements, care must be taken to insure that minimum amounts of isotopic impurity are present in the original target material and in the reagents used in the chemical separations. The precautions and techniques which are used in conventional trace analysis (12) to prevent chemical contamination are directly applicable to the problem of preventing stable isotopic contamination in the preparation of carrier-free activities. A useful method of estimating the amount of isotopic impurity contributed by the target material, reagents and vessels, is to run a "blank" on the entire separation process. Spectrographic analysis of the final product will indicate the extent of the isotopic contamination. The availability and sources of elements and compounds in high purity forms has been discussed elsewhere (11).

Although stable isotopes as well as radioactive isotopes of the desired element may be produced in the transmutation reaction, the amount of element so produced is generally small in comparison with the amount of stable isotopic material which may be added unknowingly in the separation process. This may be the case even though the isotopic impurity cannot be detected using the most sensitive analytical procedures. The only radioisotope preparations which can be obtained absolutely free from admixture with stable isotopic material are of course those of elements which do not occur naturally in detectable amounts. Although there is a certain ambiguity in the term "carrier-free" in describing radioisotope preparations of the stable elements, it is generally used, as it is in the present paper, to indicate that stable isotopic carrier materials were not added to facilitate the separation and that "reasonable" precautions were taken to insure that chemicals of maximum purity were used.

Radiochemical Contamination: Although many of the possible sources of radiochemical contamination in radioisotope preparations can be eliminated by a proper choice of target materials, bombarded energies, separation process, etc.,

radiochemical analysis of the final preparation is usually required. In determining radiochemical purity, the safest procedure is to employ both physical and chemical methods, the former based on consideration of half-life and radiation characteristics and the latter involving conventional analytical separations using added stable isotopic carriers for each of radioelements known to be produced or suspected of being produced under the particular bombardment conditions employed. Detailed procedures for the detection and identification of radioactive contaminants have been discussed in considerable detail elsewhere (23).

Target Preparations: From the standpoint of yield of radioactive product per gram of bombarded material, the most desirable target substance for both cyclotron and pile bombardment is a single isotope in the elemental form. This situation is infrequently realized, of course, because most elements are polyisotopic and the use of separated isotopes is usually not warranted in the practical production of carrier-free tracers. In addition, factors other than those based on yield considerations may be of paramount importance in determining the physical and chemical composition of target materials.

Probably the most important factor limiting the choice of cyclotron target material is the problem of dissipating the large heat input from the bombarding beam of particles. With an "internal" or "probe" target assembly, which is supported inside the vacuum chamber of the cyclotron, beam currents as high as one milliampere may be obtained over an area of approximately 0.5 cm^2 requiring a power dissipation of $\sim$40 kw/cm^2 for a 20 Mev deuteron beam. Temperature gradients in the order of 2000°C/mm may be produced under these conditions. Target materials for probe bombardments are limited to metals or metallic compounds having a high melting point and a high heat conductivity. Even for these substances the power dissipation cannot be greater than approximately 10-20 kw/cm^2. The metals are generally soldered or plated to a water-cooled support which may be made to rotate or vibrate, thus spreading the incident beam over a larger area (13).

With the "bell-jar" target assembly, a fraction of the internal beam is brought out of the cyclotron vacuum through a thin metal "window" into an external

exclosure which can be evacuated or filled with any desired gas. The intensity of the emergent beam is usually about one-tenth of the internal current. Maximum available external beam intensities are rarely used except in the bombardment of very refractory materials or in the bombardment of metals and metallic substances which have a high thermal conductivity and can be bonded to a water-cooled target plate. Additional cooling can be obtained by passing an inert gas through the target chamber. Target materials which volatilize or decompose to give a gaseous product must be irradiated with the external beam. Metal powders, oxides and other substances which are not readily bonded to the target plate are supported with metal foils - generally of platinum or tantalum. Many different target designs have been developed for special uses (18,19,20,21).

TABLE VI

OUTLINE OF METHODS FOR THE PRODUCTION
AND ISOLATION OF CARRIER-FREE RADIOISOTOPES

Radioisotope	Produced by	Description of Separation Processes
H^3	Be(d,2α) Li(n,α)	The H^3 is released by heating the Be or LiF in an evacuated quartz tube. The H^3 is condensed as H^3_2O after passing over hot CuO (55,56,57).
Be^7	Li(d,n)	Be^7 is separated from Li by co-precipitation on $Fe(OH)_3$ from dilute NH_4OH solution. The $Fe(OH)_3$ containing the Be^7 is dissolved in 6N HCl and Fe is extracted with ether (58,59).
		The Li is dissolved in H_2O. The alkaline "solution" is drawn through a sintered-glass filter which removes the Be^7 as adsorbed radiocolloid. The activity is removed from the glass surface with dilute acid (60).

Radioisotope	Produced by	Description of Separation Processes
		Be^7 is extracted from aqueous solution at pH 5-6 by thenoyl trifluoracetone in benzene. The activity is back extracted with concentrated HCl (61).
C^{11}	B(d,n)	$C^{11}O$ is expelled from a B_2O_3 target during bombardment. The $C^{11}O$ is passed over hot CuO and condensed in a liquid-air trap (62,63).
C^{14}	N(n,p)	The $C^{14}O$ and $C^{14}O_2$ formed during bombardment of NH_4NO_3 target solution are removed by aspiration with CO_2-free air. The gas is then passed over hot CuO and the $C^{14}O_2$ is collected in $Ba(OH)_2$ solution (64,65,66).
		$Ca(NO_3)_2$ target material is dissolved in a vessel filled with CO_2-free air. H_2O_2 is added to oxidize nitrogen oxides to nitrates. HNO_3 is added and the $C^{14}O_2$ is trapped in $Ba(OH)_2$ solution (67).
N^{13}	C(d,n)	The C is bombarded in a gas-tight chamber. The N^{13} released during bombardment is pumped off. The major part of the N^{13} remains in the C target material. This fraction is obtained by burning the C target in a combustion tube (68,69).
F^{18}	O(α,pn)	O_2 gas is bombarded in a gas-tight chamber. The F^{18} condenses out on the walls and is removed by washing with water (70).
	O(d,2n)	H_2O may be used as a target material to produce an aqueous solution of F^{18} directly. This target material, however, limits the beam intensity to a low value (71).
$Na^{22,24}$	Mg(d,α)	The $Na^{22,24}$ is dissolved out of MgO, $Mg(OH)_2$ or $MgCO_3$ almost quantitatively with water (72,73,74).

Radioisotope	Produced by	Description of Separation Processes
		$Na^{22,24}$ does not co-precipitate with $Mg(NH_4)_2(CO_3)_2 \cdot 4H_2O$ (75).
		A separation of Na^{22} from weighable amounts of Mg may be obtained using ion-exchange techniques (76).
Mg^{27}	Al(d,2p)	The Al target is dissolved in excess NaOH. The resultant "solution" is drawn through filter paper which separates the Mg^{27} as adsorbed radiocolloid. The activity is removed with dilute HCl (77).
P^{32}	S(n,p)	Pile bombarded S (elemental) is melted, and at a temperature of 120-130°C., is poured into boiling concentrated HNO_3 with stirring. On cooling the HNO_3 containing the P^{32} is separated from the S, and filtered. Fe^{+++} is added and the solution is made alkaline with NH_4OH. The $Fe(OH)_3$ containing the P^{32} is dissolved in HCl and Fe is extracted into ether. The P^{32} solution is then passed through a cation-exchange column (78,79).
		CS_2 may be used as target material with other neutron sources (i.e., radium-beryllium or cyclotron). The P^{32} is extracted from CS_2 by shaking with dilute HNO_3 (80,81).
		P^{32} is removed from irradiated CS_2 by the use of electrodes immersed in the liquid (82).
S^{35}	Cl(d,α) Cl(n,p)	KCl, NaCl and $FeCl_3$ are used as target materials. Separation of S^{35} from irradiated KCl on NaCl is effected by taking advantage of the insolubility of these salts in HCl. S^{35} is quantitatively retained in the HCl solution. Separation from $FeCl_3$ is effected by solvent extraction of $FeCl_3$ from HCl solutions (83,84,79).

Radioisotope	Produced by	Description of Separation Processes
		KCl is dissolved in H_2O and a few drops of H_2O_2 are added to oxidize S^{35} to $SO_4^{=}$. K^+ is removed on ion-exchange column(67).
A^{37}	Cl(d,2n)	The A^{37} is retained in the KCl target material during bombardment. Separation is made by fusing the KCl or by boiling a solution of the KCl in vacuo (85).
$K^{42,43}$	A(α,p) A(α,pn)	A stream of A is bombarded in a bell-jar target. The major portion of $K^{42,43}$ settles out on the walls of the bell-jar and the remainder is caught in a glass-wool plug in the gas outlet. After bombardment, the walls and the plug are washed in warm H_2O (86).
Ca^{45}	Sc(n,p) Sc(d,2p)	The Sc_2O_3 target is dissolved in HCl, and reprecipitated as $Sc(OH)_3$ with the addition of NH_4OH. The Ca^{45} in the supernatant is co-precipitated on $BaCO_3$. Separation from the Ba is effected by taking advantage of the insolubility of $BaCl_2$ in concentrated HCl-ether solution(87).
		The Sc_2O_3 is dissolved in 12N HCl and the solution is evaporated to incipient dryness. H_2O is added and the pH is adjusted to 4.0 with NaOH. This solution is extracted with 200 ml of 0.5 M thenoyl trifluoroacetone in benzene which removes most of the Sc. The aqueous phase containing the Ca^{45} is adjusted to pH 8 with NaOH and re-extracted with 0.5 M TTA. The Ca^{45} in the benzene fraction is back extracted with H_2O or dilute HCl (88).
$Sc^{44,46,47,48}$	Ti(d,α) (d,2p)	The Ti metal target is dissolved in a minimum volume of H_2SO_4. The solution is slowly added to excess 8N NH_4OH containing 30% H_2O_2 to give a clear "solution" of the soluble

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Radioisotope	Produced by	Description of Separation Processes
		pertitanate containing the Sc activity as radiocolloidal aggregates. This solution is then drawn through filter paper which retains over 95% of the Sc activity as adsorbed radiocolloid, which is removed by treatment with dilute HCl (89).
V^{48}	Ti(d,2n)	The TiO_2 is fused with a mixture of Na_2CO_3 and $NaNO_3$. The V^{48} is leached from the fused mass with water. The alkaline solution of V^{48} is acidified with HCl and reduced in volume to precipitate the large excess of NaCl. V^{48} is retained in the supernatant (90).
Cr^{51}	V(d,2n) V(p,n)	The V target is dissolved in HNO_3. The solution is diluted to 6N and saturated with SO_2 to insure the reduction of Cr^{+3}. Fe^{+++} or La^{+++} is added and the solution is made alkaline with Na_2CO_3. V is oxidized to soluble vanadate by air in alkaline solution and Cr^{51} is carried qualitatively on the $La(OH)_3$. Cr^{51} is separated from the $La(OH)_3$ by a second precipitation in the presence of Br_2 which oxidizes the Cr^{51} to chromate.(91)
$Mn^{52,54}$	Fe(d,α)	The Fe target is dissolved in HCl. All but a few mg of Fe is extracted with ether. The $Mn^{52,54}$ in the tetrapositive state, is quantitatively co-precipitated on $Fe(OH)_3$ with the addition of NH_4OH-Br_2 mixture. Several reprecipitations are required to obtain the $Mn^{52,54}$ free from the concurrently produced Co activities (92).
	Cr(d,n) Cr(d,2n)	The Cr target is dissolved in HCl and evaporated to incipient dryness. $FeCl_3$ carrier is added and the solution is added with stirring to NaOH solution saturated with Br_2. The $Mn^{52,54}$ is quantitatively co-precipitated on the $Fe(OH)_3$ which is

Radioisotope	Produced by	Description of Separation Processes
		dissolved in 6N HCl and extracted with ether to remove Fe carrier (93).
Fe^{59}	Co(d,2p) Co(n,p)	The Co is dissolved in 3N HNO_3 and the pH of the solution is adjusted to 4.0-7.0 by the addition of NH_4OH-NH_4Ac. 1 ml of a saturated aqueous solution of acetyl acetone is added and the solution is extracted with xylene. The organic phase containing the Fe^{59} is evaporated to dryness. Organic residue is destroyed by fuming $HClO_4$ (94).
$Co^{55,56,57,58}$	Fe(d,xn)	The Fe target is dissolved in HCl. $Fe(OH)_3$ is precipitated with the addition of excess NH_4OH-NH_4Cl solution containing Br_2. The Co^{56} is retained in the supernatant. NH_4 salts are destroyed with HNO_3 (92).
$Cu^{64,67}$	Zn(d,2p)	The Zn target is dissolved in HCl, diluted to 5.5N and Ga activities are extracted with ether to reduce radiation hazard in subsequent manipulations. The pH of the aqueous phase is adjusted to 1.0-1.2 with NaOH and the Cu^{64} is extracted with CCl_4 solution containing .001% dithizone. The CCl_4 phase is evaporated to dryness and organic material is removed by heating to 500°C (95,96).
	Zn(n,p)	$ZnSO_4$ target is dissolved in water. The Cu^{64} is separated from the acidified solution by shaking the solution with a few milligrams of freshly precipitated Bi_2S_3, dissolving the Bi_2S_3 with HNO_3, evaporating the solution to dryness, taking up in dilute HCl and reprecipitating the Bi as $Bi(OH)_3$ with excess NH_4OH. The Cu^{64} is retained in the supernatant (97).

Radioisotope	Produced by	Description of Separation Processes
		$ZnCl_2$ target is dissolved in water. Cu^{64} is concentrated by addition of small amount of Zn which is then dissolved in acid and Cu^{64} is deposited on polished Pt foil from 0.05N H_2SO_4 saturated with H_2 (98).
Zn^{65}	Cu(d,2n)	Cu target is dissolved in HNO_3. Excess HCl is added to destroy HNO_3 and the solution is neutralized to 0.25N HCl with NH_4OH. CuS is precipitated with H_2S. Zn^{65} is not co-precipitated. The supernatant is adjusted to pH 5-5.5 with NaAc and sufficient $Na_2S_2O_3$ is added to complex traces of Cu remaining in solution. The Zn^{65} is extracted into CCl_4 solution containing dithizone and back extracted with .02N HCl (99).
Ga^{67}	Zn(d,n) (d,2n)	Zn target is dissolved in HCl and adjusted to 6N. Ga^{67} is extracted with ether (100).
Ge^{71}	Ga(d,2n)	The Ga target is dissolved in 48% HBr and the solution is distilled. The Ge^{71} is obtained in the first few ml of the HBr distillate. HNO_3 is added to destroy HBr and the solution is evaporated to incipient dryness. H_2O is added and the evaporation is repeated to remove traces of HNO_3 (141).
As^{74}	Ge(d,2n)	The Ge metal is dissolved by refluxing with aqua regia. Excess HCl is added to destroy HNO_3 and $GeCl_4$ is distilled from the solution. The As^{74} is retained in the residue as the non-volatile pentachloride. After $GeCl_4$ is completely removed, HBr is added to the HCl residue and As^{74}, as the volatile trichloride, is distilled into HNO_3. The HNO_3 solution of As^{74} is evaporated to dryness (101,102).

Radioisotopes	Produced by	Description of Separation Processes
Se^{75}	As(d,2n)	The As is dissolved in aqua regia. HCl is added to destroy excess HNO_3. The solution is adjusted to 3N. Tellurous acid carrier is added and precipitated as Te by passing SO_2 through the solution. Se^{75} is carried quantitatively. The Te precipitate is dissolved in HNO_3. HBr is added and the Te is distilled. Se^{75} is retained in the HNO_3 residue (103).
$Br^{80,82}$	Se(d,2n)	The $Br^{80,82}$ is volatilized while the Se target is dissolved in H_2SO_4. An inert gas is used to sweep the $Br^{80,82}$ into a trap filled with CCl_4. A major fraction of the activity is extractable with Na_2SO_3 solution. Some of the $Br^{80,82}$ reacts with an unidentified impurity in the CCl_4 (104).
K^{79}	Br(d,2n)	The K^{79} is retained in the KBr target material during bombardment. Separation is made by fusing the KBr or by boiling a solution of the KBr in vacuo (105,106).
Sr^{85}	Rb(d,2n)	The RbCl is dissolved in 0.1N HCl, $FeCl_3$ carrier is added and precipitated with the addition of $(NH_4)_2CO_3$-NH_4OH solution. The $Fe(OH)_3$ containing the Sr^{85} activity is dissolved in 6N HCl and Fe is extracted with ether (107).
$Sr^{89,90}$	U(n,f)	$Sr^{89,90}$ is separated from fission-product mixture by complex elution from organic zeolites. $Sr^{89,90}$ and Ba^{140} are removed successively by 5% citrate at pH 5 (108,109). (A more detailed description of fission-product separation by ion-exchange methods is given in the section on rare-earths.)

Radioisotopes	Produced by	Description of Separation Processes
Y^{88}	Sr(d,2n)	The Sr metal is dissolved in dilute HCl. The solution is diluted to ~0.5 M Sr^{++} and NH_4OH is added to pH 9. This "solution" is then drawn through filter paper which removes the Y^{88} as adsorbed radiocolloid. The activity is removed with dilute HCl (110).
Y^{91}	U(n,f)	Y^{91} is separated from rare-earth fission products by complex elution from amberlite IR-1 resin with 5% citrate at pH 2.7 (108,111). (A more detailed description of fission-product separations is given in the section on rare earths.)
Zr^{89}	Y(d,2n)	The Y_2O_3 target is dissolved in HCl. The resultant solution is evaporated almost to dryness and then diluted with H_2O. Y is precipitated as the fluoride with HF. A few ml of H_2SO_4 are added and HF is removed by evaporation. The solution is diluted with water, $FeCl_3$ carrier is added and precipitated with NH_4OH. The Zr^{89} is carried quantitatively. The precipitate is dissolved in 6N HCl and Fe is extracted with H_2O(107).
Zr^{95}	U(n,f)	Oxalic acid is used as a specific complexing agent to remove Zr^{95} and Cb^{95} from mixed fission products on amberlite IR-1 columns - 0.5% oxalic strips Zr^{95}-Cb^{95} quantitatively; none of the trivalent or divalent elements is removed by this treatment (108).
		Zr^{95} is separated from Cb^{95} daughter by, (a) co-precipitation of Cb^{95} on MnO_2 from 10N HNO_3, and (b) extraction of Zr^{95} from HNO_3 or $HClO_4$ using thenoyl trifluoroacetone (112).

Radioisotopes	Produced by	Description of Separation Processes
		The bulk of the uranyl nitrate target material is removed by ether extraction. To the concentrated solution of fission products are added a few mg. of $FeCl_3$, hydroxylamine and excess NH_4OH. Zr^{95} and other NH_3 insoluble fission products are co-precipitated on the $Fe(OH)_3$. The U remains in the supernatant. The $Fe(OH)_3$ is dissolved in HCl and Fe is extracted with ether. The Zr^{95}, Cb^{95} and $Ce^{141,144}$ are separated as iodates using thorium carrier. Zr^{95}-Cb^{95} are separated from Ce and Th by means of HF precipitation. Cb^{95} is separated from the Zr^{95} by K_2CO_3 fusion (107).
Cb^{95}	U(n,f)	See Zr^{95} procedures.
Mo^{99}	Zr(α,n)	The ZrO_2 target is dissolved in the presence of Cb "hold-back" carrier by boiling in 12N H_2SO_4 down to fuming. NaCl and HCl are added, following cooling and dilution with water, to bring the (H^+) and (Cl^-) to 6N. The Mo is extracted with ether saturated with HCl (113).
Tc^{95}	Mo(d,n)	The Mo target is dissolved in aqua regia and evaporated to incipient dryness. The oxide is dissolved in $HClO_4$-H_3PO_4 and Tc^{95} is distilled with the addition of HBr using a CO_2 carrier gas (114,115).
Tc^{99}	U(n,f)	The U metal is dissolved in 37% HCl and the UCl_4 is oxidized to UO_2Cl_2 with H_2O_2 and Br_2 at 60°C. $PtCl_2$ is added and the solution is saturated with the H_2S at 90°C. The Tc^{99} is co-precipitated with the PtS_2 along with other acid insoluble sulfides of the fission products. The PtS_2 is dissolved in H_2O_2-NH_4OH solution, Br_2 is added and the solution is

Radioisotopes	Produced by	Description of Separation Processes
		evaporated to incipient dryness. 18N H_2SO_4 is added and the Tc^{99} is distilled (116).
Ru^{103}	U(n,f)	The U metal is dissolved in HNO_3 and adjusted to 3N. NaI is added and the solution is boiled until all of the iodine is expelled. $HClO_4$ is added and Ru^{103} is distilled into 3N HCl. The distillate is made strongly alkaline and Ru^{103} is distilled from alkaline solution after oxidation with Cl_2(107).
Pd^{103}	Rh(d,2n)	The Rh is fused with $KHSO_4$ and the fused mass is dissolved in water. HCl is added together with mg amounts of H_2SeO_4 and the solution is saturated with SO_2. The Pd^{103} is carried quantitatively on the Se metal. Se is removed by distillation with $HClO_4$-HBr. (117)
$Ag^{105,106,111}$	Pd(d,n) (d,2n)	Ag^{105} is separated by co-precipitation on Hg_2Cl_2 which is removed by volatilization (118)
Ag^{111}	Pd^{111} β decay	Ag^{111} is separated from Pd by electrolysis from 1 M NaOH-0.1 M NaCN solution using Pt electrodes and a cathode potential of -1.210 V (119).
Cd^{109}	Ag(d,2n)	The Ag is dissolved in HNO_3 and the solution is evaporated to dryness. The $AgNO_3$ containing the activity is dissolved in H_2O and the Ag^+ is complexed with NH_4CNS. The solution is adjusted to pH 5 with sodium acetate and the Cd^{109} is extracted with chloroform containing 5% pyridine (120).

Radioisotopes	Produced by	Description of Separation Processes
$In^{111,114}$	Cd(α,p) (α,pn)	After separation of Sn^{113} by distillation (see below), the H_2SO_4 residue containing In^{114} is neutralized with NH_4OH. The $Fe(OH)_3$ precipitate carries the In^{114} quantitatively. Fe is separated by ether extraction (121).
Sn^{113}	Cd(α,n) (α,2n)	The Cd is dissolved in HNO_3 and the solution is evaporated to incipient dryness, diluted with H_2O and made alkaline after the addition of $FeCl_3$. The Sn^{113} and In^{114} are carried on the $Fe(OH)_3$ precipitate. The $Fe(OH)_3$ is dissolved in a minimum volume of 36N H_2SO_4 and transferred to an all-glass distilling apparatus. HBr is added dropwise and Sn^{113} is volatilized at 220°C. The distillate is collected in HCl(121).
$Sb^{120,122}$	Sn(d,n) (d,2n)	The Sn target is dissolved in aqua regia. HCl is added to destroy excess HNO_3 and the solution is adjusted to 0.1N HCl. Milligram amounts of Cd^{++} are added and precipitated with H_2S after the addition of oxalic acid to prevent the precipitation of SnS_2. The CdS is dissolved in HCl and transferred to a distilling flask. Traces of Sn are removed by distillation with $HClO_4$ at 200°C. $Sb^{120,122}$ is then distilled with the gradual addition of HBr (121).
$Te^{127,129}$	U(n,f)	The $Te^{127,129}$ is precipitated out of an HCl fission mixture on CuS. The CuS is dissolved in HNO_3 and the solution is made 5N in HF. The $Te^{127,129}$ is then co-precipitated on RuS_2. The RuS_2 is decomposed with HNO_3 and the residue is boiled with $HClO_4$ to expel the Ru (107).

Radioisotopes	Produced by	Description of Separation Processes
I^{131}	Te(d,n) (d,2n)	The Te is dissolved in CrO_3-H_2SO_4 solution. Oxalic acid is added and the I* is distilled into NaOH (123). *I is evolved during the dissolution of Te target in HNO_3. The activity is collected in a trap filled with CCl_4 and extracted with Na_2SO_3 (107).
	U(n,f)	I^{131} vaporizes during the solution of U in HNO_3. The vapor is passed through a condenser and NaOH scrubber. I^{131} is redistilled into dilute NaOH from HNO_3-H_2O_2. The NaOH solution of I^{131} is made 20% in H_2SO_4 and treated with $KMnO_4$. Oxides of nitrogen are removed by distillation. The residue is treated with H_3PO_3 and the I^{131} is distilled into dilute NaOH-Na_2SO_3 solution (124,67).
	Te^{131} β^- decay	The Te metal is fused with NaOH. The fused mass is extracted with water and the washings are transferred to an all-glass distilling flask. $KMnO_4$ is added to the alkaline solution to oxidize I^{131} to IO_3^-. H_2SO_4 is then added and the I^{131} is distilled into dilute Na_2SO_3-Na_2CO_3 solution (125).
		Te target is dissolved in 18N H_2SO_4-50% CuO_3 solution. The solution is cooled to 50°C and solid oxalic acid is slowly added. The I^{131} is then distilled into dilute Na_2SO_3-NaOH solution (124,67).
Xe^{127}	I(d,2n)	The Xe^{127} is retained in the KI target material during bombardment. Separation is made by fusing the KI or boiling a solution of the KI in vacuo (107,85).

Radioisotopes	Produced by	Description of Separation Processes
Cs^{131}	Ba^{131} β^- decay	$BaCl_2$ is precipitated from 12N HCl. Cs^{131} remains in the supernatant. Further purification is obtained by $La(OH)_3$ scavenging (126).
Cs^{137}	U(n,f)	Cs^{137} is separated from the divalent or trivalent fission product ions by preferential elution from amberlite IR-1 using 0.25N HCl (127).
		Cs^{137} is separated from fission mixture by co-precipitation on NH_4ClO_4 from $HClO_4$ solutions using absolute alcohol(128).
		Cs^{137} is separated from a fission product mixture by co-precipitation on sodium cobaltinitrite and ammonium cobaltinitrite. The Co is removed as CoS from NH_4OH solution (107).
Ba^{133}	Ca(d,2n)	The CsCl target material is dissolved in HCl to pH 2 and the solution is adjusted to pH 10 with NH_4OH after the addition of $FeCl_3$ carrier. The precipitate of $Fe(OH)_3$ is washed, dissolved in HCl and reprecipitated with NH_4OH at pH 5.4. The Ba^{133} is retained in the supernatant (129,107).
Ba^{140}	U(n,f)	After the elution of the rare-earths from Amberlite IR-1 using 5% citrate at pH 2.7, the $Sr^{89,90}$ and Ba^{140} are successively removed by 5% citrate at pH 5 (108,130).
La^{140}	Ba^{140} β^- Decay	La^{140} is separated from Ba^{140}-La^{140} mixtures by elution from Amberlite IR-1 using 5% citrate at pH 2.7 (108,130).
		$BaCl_2$ is added to an HCl solution of mixed fission products and precipitated by passing HCl gas through the solution. The $BaCl_2$ containing the Ba^{140} is dissolved in H_2O. $FeCl_3$ carrier

Radioisotopes	Produced by	Description of Separation Processes
		is added and precipitated with the addition of NH_4OH. The $Fe(OH)_3$ precipitate is discarded. After allowing the purified $BaCl_2$ to stand for several days a second $Fe(OH)_3$ precipitation is made which carries the La^{140}. Fe is removed by ether extraction from 6N HCl (107).
58-63		
$Ce^{141,144}$ Pr^{143} Nd^{147} Pm^{147} Sm^{151} Eu^{155}	U(n,f)	A solution of all fission-produced species in 0.01 to 0.1N HCl is passed through an Amberlite IR-1 resin column. All cations are adsorbed while the fission produced anionic elements (Ru, Te, Tc) pass through acid are recovered in the effluent. Most of the Cs^{137} may be eluted at this point by the addition of 0.25N HCl. The column is washed, and Cb^{95}-Zr^{95} is preferentially removed by 0.5% oxalic acid. By using differential pH - controlled ammonium citrate elution, the trivalent elements are removed in the order: Y, Eu, Sm, Pm, Nd, Pr, Ce, La. With Amberlite IR-1, the eluting agent in general use is 4.75-5.0% citric acid at a pH of from 2.7-2.95. In practice a group of several elements may be eluted then readsorbed and fractionated under more rigidly controlled conditions (108,127,67).
64-71 Gd Tb Dy Ho Er Tm Yb	Z(p,xn) (d,xn) (α,xn)	After bombardment, the rare earth oxide is dissolved in HNO_3 and insoluble matter is removed by centrifugation. Carriers for contaminating activities are added and the rare earth fluoride is precipitated from hot 2N HNO_3 by addition of HF. The fluoride is dissolved in HNO_3-H_3BO_3 and precipitated as the hydroxide. The rare-earth hydroxide is dissolved in HCl and the solution is adjusted to a pH between 0.5 and 1.5. The rare earth activities are then adsorbed onto Dowex-50 and

Radioisotopes	Produced by	Description of Separation Processes
		eluted with 0.25 M citric acid adjusted to pH 3.05 with NH_4OH (131).
$Ta^{177,178,179}$	Hf(d,xn)	The HfO_2 is dissolved in HNO_3-HF solution. HF is removed by evaporation with the addition of 16N HNO_3. $KMnO_4$ is added and the MnO_2 precipitate carries the Ta quantitatively. The MnO_2 containing the Ta activity is dissolved in oxalic acid and Mn^{++} is removed by adsorption on Amberlite IR-1 or Dowex 50 (132).
$Re^{183,184}$	Ta(α,xn) W(d,xn)	The Ta metal target is dissolved in a minimum volume of 16N HNO_3 containing 10% HF. The HF is removed by volatilization and the bulk of the tantalic acid is separated by centrifugation. The $Re^{183,184}$ is quantitatively retained in the supernatant. The HNO_3 solution is evaporated almost to dryness and transferred to an all-glass distilling flask with 36N H_2SO_4. The $Re^{183,184}$ is distilled at 240°C with the addition of 9N HBr and co-precipitated on CuS after the removal of HBr and HNO_3. Separation from the Cu is effected by precipitating CuO from alkaline solution (133,134).
Os^{185}	W(α,xn)	The W metal is fused with KOH-KNO_3 and the fused mass is extracted with water. The solution is acidified with HNO_3 precipitating most of the WO_3 which is removed by centrifugation. The Os^{185} is distilled from 5N HNO_3 and collected in trap filled with 5N HNO_3. Extraction of the HNO_3 distillate with ether removes the Os^{185} which is back extracted with NaOH(135).

Radioisotopes	Produced by	Description of Separation Processes
$Bi^{204,206}$	Pb(d,2n)	The Pb is dissolved in a minimum volume of HNO_3 and the resultant solution is evaporated to dryness. The $Pb(NO_3)_2$ containing the $Bi^{204,206}$ is dissolved in excess 10% NaOH. This "solution" is drawn through filter paper which retains over 98% of the Bi activity as adsorbed radiocolloid. Dilute HCl removes the $Bi^{204,206}$ quantitatively (136).
Po^{210}	Pb(α,xn)	The Pb target is dissolved in HNO_3 $Pb(NO_3)_2$ is centrifuged off and the solution is extracted with amyl acetate to remove Tl, Hg and Au. Bi and Tl hold back carriers are added and the solution is fumed with HCl. The solution is diluted to 3N and Te carrier is precipitated with SO_2 removing At and Po. The Te is reprecipitated from 6N HCl. Under these conditions At is co-precipitated and Po remains in the supernatant. Po in the HCl solution is extracted with an equal volume of 20% tributyl phosphate in diethyl ether. The activity is back extracted with HNO_3 (137).
Po^{210}	Bi(d,n)	Same procedure as above (137).
$At^{210,211}$	Bi(α,3n) Bi(α,2n)	A layer of Bi metal alloyed to a 10 mil thick water-cooled Al target plate is bombarded in a bell-jar target filled with He for additional cooling and to prevent oxidation of the Bi. The bombarded Bi is cut from the target foil and At is isolated by heating the Bi to 425°C. in a stream of N_2 carrier-gas. The At is collected on a cold-finger cooled with liquid air (138,139).

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