PRODUCTION AND ISOLATION OF CARRIER-FREE RADIOISOTOPES[1]

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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.[2]

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[2] Nontransmutation 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 atom by breaking its chemical bond in the target

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 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 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.

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or parent compound as a result of isomeric transition (29, 117) or recoil (85, 123). These methods, although of considerable theoretical interest, are at present not generally practical for the production of carrier-free tracers.

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 the functions of trace elements, 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.

A. 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 (103, 116). 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 1 week are described in table 1. Most of these are available in carrier-free form from the Isotopes Division, Atomic Energy Commission, Oak Ridge, Tennessee (64).

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

TABLE 1

Major radioactive fission products of half-life greater than 1 week (64, 103, 116)

RADIOISOTOPE	HALF-LIFE	ENERGY		RELATIVE YIELD*
		Beta	Gamma	
		M.e.v.	*M.e.v.*	*curies*
$_{38}Sr^{89}$	55 days	1.48	None	1.0
$_{38}Sr^{90}$†	25 yr.	0.65	None	0.035
$_{39}Y^{91}$	57 days	1.53	None	1.25
$_{40}Zr^{95}$	65 days	0.394 (98%)	0.73 (93%)	1.35
$_{41}Nb^{95}$	35 days	0.154	0.75	1.7
$_{43}Tc^{99}$	$\sim 5 \times 10^5$ yr.	0.32	None	0.7×10^{-6}
$_{44}Ru^{103}$	42 days	0.2 (95%)	0.56	0.9
$_{44}Ru^{106}$	1.0 yr.	0.04	None	0.065
$_{52}Te^{127m}$†	90 days	I.T., e^-	0.086	0.0075
$_{52}Te^{129m}$	32 days	I.T., e^-	0.102	0.03
$_{53}I^{131}$	8 days	0.35; 0.60	0.63; 0.36; 0.28	0.065
$_{55}Cs^{137}$	33 yr.	0.55; 0.84	0.66	0.032
$_{56}Ba^{140}$†	12.5 days	1.05	0.529	0.35
$_{58}Ce^{141}$	28 days	0.66	0.2	1.0
$_{58}Ce^{144}$†	275 days	0.35	None	0.85
$_{59}Pr^{143}$	13.8 days	0.83	None	0.4
$_{60}Nd^{147}$	11 days	0.90	0.55	0.12
$_{61}Pm^{147}$	3.7 yr.	0.22	None	0.14

* Relative amounts of fission products coexisting one month from the end of a several-month exposure (103).

† Indicates that the radioisotope decays to a more energetic daughter (103, 116).

be used to produce carrier-free radioisotopes if the product isotope decays to give a radioactive descendant.

Table 2 describes the carrier-free radioisotopes which are routinely produced in the pile using this type of nuclear reaction. The activation cross-section of

TABLE 2

Carrier-free radioisotopes produced in the pile by (n,γ) reaction followed by decay (64, 116, 119)

RADIOISOTOPE	HALF-LIFE	TARGET ISOTOPE	PER CENT ABUNDANCE	THERMAL NEUTRON ACTIVATION CROSS-SECTION (BARNS)		HALF-LIFE OF (n,γ) PRODUCT
				Isotope	Natural	
$_{33}As^{77}$	40 hr.	Ge^{76}	6.5	0.085	0.0055	12 hr.
$_{41}Nb^{95}$	35 days	Zr^{94}	17.4	0.06	0.014	65 days
$_{43}Tc^{97m}$	93 days	Ru^{96}	5.68	2.2	0.15	2.8 days
$_{43}Tc^{99}$	$\sim 5 \times 10^5$ yr.	Mo^{98}	24.1	0.415	0.10	67 hr.
$_{45}Rh^{105}$	37 hr.	Ru^{104}	18.27	0.667	0.122	4.4 hr.
$_{47}Ag^{111}$	7.5 days	Pd^{110}	13.5	0.39	0.053	26 months
$_{51}Sb^{125}$	2.7 yr.	Sn^{124}	6.8	0.574	0.039	9 months
$_{53}I^{131}$	8.0 days	Te^{130}	34.52	0.22	0.0735	25 months
$_{55}Cs^{131}$	10.2 days	Ba^{130}	0.101			12.0 days
$_{59}Pr^{148}$	13.8 days	Ce^{142}	11.7	0.95	0.11	33 hr.
$_{61}Pm^{147}$	3.7 yr.	Nd^{146}	17.1	1.4	0.24	11.0 days
$_{61}Pm^{149}$	47 hr.	Nd^{148}	5.78	2.8	0.16	1.7 hr.
$_{63}Eu^{155}$	2 yr.	Sm^{154}	22.53	5.50	1.10	25 months
$_{79}Au^{199}$	3.3 days	Pt^{198}	7.2	3.92	0.28	31 months
$_{84}Po^{210}$	140 days	Bi^{209}	100	0.015	0.015	5.0 days

Carrier-free radioisotopes produced in the pile by (n,p) and (n,α) reaction

RADIOISOTOPE	HALF-LIFE	PRODUCED BY	REFERENCES
$_1H^3$	12 yr.	Li^6 (n,α)	(97)
$_6C^{14}$	5100 yr.	N^{14} (n,p)	(96, 108, 134)
$_{15}P^{32}$	14.3 days	S^{32} (n,p)	(16, 30, 32, 47, 114)
$^{16}S^{35}$	87.1 days	Cl^{35} (n,p)	(68, 110, 114, 131)
$_{20}Ca^{45}$	180 days	Sc^{45} (n,p)	(9, 72)

the target isotope and the characteristics of the radioactive descendant are also listed.

3. Neutron capture with proton and α-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 charged-particle emission above $Z = 20$. Resonance in certain nuclei also permits (n,p) and (n,α) reactions to proceed with neutrons of low energy. Table 2 lists the radioisotopes which are produced by (n,p) and (n,α) reaction in the pile.

B. CYCLOTRON REACTIONS

The cyclotron is the only practical source of a large number of useful carrier-free radioisotopes which are not available from pile sources (64, 116). In addition, although a desired radioisotope may be produced in the pile, half-life and other technical limitations may make cyclotron irradiation the more desirable 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-M.e.v. deuterons, for example, three and occasionally four radioactive elements may be produced concurrently from the target element (130). At energies considerably higher than this ($\sim$100 M.e.v.) a whole spectrum of transmutation products is 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 (25, 46, 86). 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. The intensity of the charged particles is low at these energies and the radiochemical procedures become formidable, owing to the large number of radioelements produced.

Because of the great variety of nuclear transmutation reactions (115, 116) which can be brought about by bombardment with deuterons, protons, and alpha particles, 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.

1. Deuteron reactions

For the production of many useful radioisotopes in the carrier-free state the reaction types (d,α), $(d,2p)$, (d,n), and $(d,2n)$ 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 $(d,\alpha p)$ 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 nontransmutative (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 nucleus. 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 (14). Nuclear reactions for which excitation functions (σ *versus* E) have been determined are listed with references in table 3. 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 (14), although usually they are obtained directly from thick-target bombardments. Available thick-target yields for charged-particle reactions are given in table 4.

2. Alpha-particle (helium-ion) reactions

Alpha-particle capture followed by neutron emission—$(\alpha,n)(\alpha,2n)(\alpha,3n)$ 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, in the preparation of radioisotopes of astatine ($Z = 85$), because the only target element available 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 3. Thick-target yields are given in table 4.

3. 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 higher energies are more readily obtained. Yield data for a few proton reactions are given in tables 3 and 4.

TABLE 3

Excitation functions of transmutation reactions

Table of references

TARGET (NATURAL ELEMENT)	REFERENCE	TRANSMUTATION REACTIONS	MAXIMUM ENERGY
			M.e.v.
C	(94)	$C(d,n)N^{13}$	5
N	(3)	$N(n,p)C^{14}$; $N(n,\alpha)B^{11}$	1.7
O	(28)	$O(p,n)F^{18}$	4
Mg	(60)	$Mg(d,\alpha)Na^{24}$	3.5
	(76)	$Mg(d,\alpha)Na^{22,24}$	14
Al	(13)	$Al(d,\alpha p)Na^{24}$	14
S	(73)	$S(n,p)P^{32}$	5.8
Cl	(107)	$Cl(n,p)S^{35}$	14
	(107)	$Cl(d,\alpha)S^{35}$	14
Fe	(20)	$Fe(d,n)Co^{55}$	10
	(21)	$Fe(d,n)Co^{55}$; $Fe(d,\alpha)Mn^{52}$	10
Ni	(122)	$Ni(p,n)Cu^{61,62,64}$	6.3
	(26)	$Ni(p,n)Cu^{64}$	4
	(126)	$Ni(d,n)Cu^{61}$	5
Cu	(122)	$Cu(p,n)Zn^{63}$	6.3
	(88)	$Cu(d,2n)Zn^{63}$	16
	(26)	$Cu(p,n)Zn^{63}$	4
	(15)	$Cu(d,2n)Zn^{63,65}$; $Cu(d,\alpha)Ni^{63}$	14
Zn	(11)	$Zn(p,n)Ga^{64,68,70}$	6.3
	(28)	$Zn(p,n)Ga^{68,70}$	4
Se	(11)	$Se(p,n)Br^{80,82}$	6.3
	(75)	$Se(p,n)Br^{80,82}$	4.0
Br	(14)	$Br(d,2n)Kr^{79}$	13.5
Rh	(8)	$Rh(\alpha,n)Ag^{106}$; $Rh(\alpha,2n)Ag^{105}$	19
Pd	(74)	$Pd(d,n)Ag^{106}$; $Pd(d,2n)Ag^{106}$	6
Ag	(75)	$Ag(d,2n)Cd^{107,109}$	9
	(8)	$Ag(\alpha,n)In^{112}$; $Ag(\alpha,2n)In^{111}$	19
	(38)	$Ag(\alpha,xn)In^{109,110,111}$	37
In	(125)	$In(\alpha,n)Sb^{118}$; $In(\alpha,2n)Sb^{117}$; $In(\alpha,3n)Sb^{116}$	38
Au	(76)	$Au(d,2n)Hg^{197}$	9
Tl	(77)	$Tl(d,n)Pb^{206}$	9
Pb	(33)	$Pb(d,n)Bi^{207}$	9
Bi	(62)	$Bi(d,n)Po^{210}$	9
	(77)	$Bi(d,n)Po^{210}$	9
	(69)	$Bi(d,n)Po^{210}$; $Bi(d,3n)Po^{208}$	19
	(19)	$Bi(d,n)Po^{210}$	14.5
	(125)	$Bi(\alpha,2n)At^{211}$; $Bi(\alpha,3n)At^{210}$	38

TABLE 4

Thick-target yields of some artificial radioelements (52)

Yields are given in terms of microcuries per microampere hour. 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, with the exception of beryllium, whose yield values are enclosed in parentheses, since they are all substances decaying by orbital electron 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 1 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 per cent 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 M.e.v., and helium ions at 38 M.e.v. 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 ±25 per cent. Those not underlined are subject to much greater error.

ISOTOPE	HALF-LIFE	RADIATIONS	REACTION	YIELD IN MICROCURIES PER MICROAMPERE HOUR				ISOTOPE RATIO
				8-M.e.v. deuterons	14-M.e.v. deuterons	19-M.e.v. deuterons	38-M.e.v. α-particles	
$_1H^3$.....	12.4 yr.	β^-	$Be^9(d,t)$		0.1			1
$_4Be^7$....	52.9 days	K, γ	$Li^7(d,2n)$			2		1.1
$_6C^{11}$....	21 months	β^+	$B^{11}(d,n)$	500				5
$_6C^{14}$....	~ 5100 yr.	β^-	$N^{14}(n,p)$		0.0005*			1
$_7N^{13}$....	10 months	β^+	$C^{12}(d,n)$	1000				1
$_9F^{18}$....	112 months	β^+	$O^{16}(\alpha,pn)$				5000	1
$_{11}Na^{22}$..	2.6 yr.	β^+, γ	$Mg^{24}(d,\alpha)$		1.0			9
$_{11}Na^{24}$..	14.9 hr.	β^-, γ	$Na^{23}(d,p)$			1500		1
$_{12}Mg^{27}$..	10.2 months	β^-, γ	$Mg^{26}(d,p)$	500	2800			9
$_{12}Mg^{27}$..	10.2 months	β^-, γ	$Al^{27}(d,2p)$			2500		1
$_{15}P^{32}$....	14.1 days	β^-	$P^{32}(d,p)$	30	120			1
$_{16}S^{35}$....	87 days	β^-	$Cl^{37}(d,\alpha)$			8.4		4

* Calculated from known neutron yields and ρ_a for nitrogen in 100 gallons of a saturated solution of ammonium nitrate.

TABLE 4—*Continued*

ISOTOPE	HALF-LIFE	RADIATIONS	REACTION	YIELD IN MICROCURIES PER MICROAMPERE HOUR				ISOTOPE RATIO
				8-M.e.v. deuterons	14-M.e.v. deuterons	19-M-e.v. deuterons	38-M.e.v. α particles	
$_{17}Cl^{38}$...	38 months	β^-, γ	$Cl^{37}(d,p)$	1000				4
$_{19}K^{42}$...	12.4 hr.	β^-, γ	$K^{41}(d,p)$		50	107		15
$_{19}K^{42}$...	12.4 hr.	β^-, γ	$Ca^{44}(d,\alpha)$			3.3		50
$_{19}K^{42}$...	12.4 hr.	β^-, γ	$A^{40}(\alpha,pn)$				2000	1
$_{19}K^{43}$...	22.4 hr.	β^-, γ	$A^{40}(\alpha,p)$				1000	1
$_{20}Ca^{45}$...	152 days	β^-	$Ca^{44}(d,p)$		0.01	0.05		50
$_{21}Sc^{46}$...	85 days	β^-, γ	$Ti^{48}(d,\alpha)$			0.01		1.3
$_{24}Cr^{51}$...	26 days	K, e^-, γ	$V^{51}(d,2n)$			(0.2)		1
$_{25}Mn^{52}$...	6.5 days	K, β^+, γ	$Cr^{52}(d,2n)$			(8)		1.2
$_{25}Mn^{54}$...	310 days	K, γ	$Fe^{56}(d,\alpha)$		(0.10)			1.1
$_{26}Fe^{59}$...	46 days	β^-, γ	$Fe^{58}(d,p)$		0.03			360
$_{26}Fe^{55}$...	4 yr.	K	$Mn^{55}(d,2n)$			(0.02)		17
$_{27}Co^{57}$...	270 days	K, β^+, e^-, γ	$Fe^{56}(d,n)$		(1.0)	(5.0)		1.1
$_{29}Cu^{64}$...	12.9 hr.	K, β^-, β^+, γ	$Cu^{63}(d,p)$		(3000)			1.5
$_{29}Cu^{64}$...	12.9 hr.	K, β^-, β^+, γ	$Zn^{64}(d,2p)$			(300)		2.1
$_{29}Cu^{67}$...	60 hr.	β^-	$Zn^{67}(d,2p)$			10		25
$_{30}Zn^{65}$...	250 days	K, β^+, e^-, γ	$Cu^{65}(d,2n)$		(0.5)			3
$_{31}Ga^{67}$...	78 hr.	K, e^-, γ	$Zn^{66}(d,n)$			(30)		3.8
$_{32}Ge^{71}$...	11.4 days	K, e^-, γ	$Ga^{71}(d,2n)$			(8)		2.5
$_{33}As^{74}$...	17.5 days	β^+, β^-, γ	$Ge^{74}(d,n)$		2	10		3

TABLE 4—*Continued*

ISOTOPE	HALF-LIFE	RADIATIONS	REACTION	YIELD IN MICROCURIES PER MICROAMPERE HOUR				ISOTOPE RATIO
				8-M.e.v. deuterons	14-M.e.v. deuterons	19-M.e.v. deuterons	38-M.e.v. α-particles	
$_{34}Se^{75}$...	127 days	K, e^-, γ	$As^{75}(d, 2n)$			(1)		1
$_{35}Br^{82}$...	35 hr.	β^-, γ	$Se^{82}(d, 2n)$				500	10
$_{37}Rb^{86}$..	19.5 days	β^-, γ	$Sr^{88}(d,\alpha)$		1.0			1.2
$_{38}Sr^{85}$...	65 days	K, γ	$Rb^{85}(d, 2n)$		(0.13)	(0.60)		1.2
$_{38}Sr^{89}$...	54 days	β^-	$Sr^{88}(d,p)$		10.4			1.2
$_{39}Y^{88}$....	105 days	K, β^+, γ	$Sr^{88}(d,2n)$		(0.10)	(1.0)		1.2
$_{40}Zr^{89}$...	78 hr.	β^+	$Y^{89}(d,2n)$		7.0	75.0		1
$_{40}Zr^{95}$	65 days	β^-, e^-, γ	$Zr^{94}(d,p)$		0.15			5.9
$_{41}Nb^{90}$..	16 hr.	β^+, γ	$Mo^{92}(d, \alpha)$		2.4			6.2
$_{41}Nb^{91}$..	55 days	K, e^-, γ	$Zr^{90}(d,n)$		(1.0)			2
$_{41}Nb^{95}$..	37 days	β^-, e^-, γ	$Mo^{97}(d,\alpha)$		0.05			11
$_{42}Mo^{99}$..	67 hr.	β^-, γ	$Zr^{96}(\alpha,n)$				0.1	36
$_{44}Ru^{97}$..	2.8 days	K, e^-, γ	$Mo(\alpha,xn)$				(30)	†
$_{45}Rh^{101}$..	4.3 days	K, e^-, γ	$Ru^{100}(d, 2n)$			(10)		7.9
$_{46}Pd^{103}$..	17 days	K	$Rh^{103}(d, 2n)$			(0.05)		1
$_{47}Ag^{110}$..	40 days	K, e^-, γ	$Pd^{110}(d, 2n)$			(0.10)		7.4
$_{48}Cd^{109}$..	470 days	K	$Ag^{109}(d, 2n)$			(2)		2.1
$_{49}In^{111}$..	2.8 days	K, e^-, γ	$Cd(\alpha,pxn)$				(0.10)	†
$_{49}In^{114m}$.	50 days	I.T., e^-						

† Difficult to evaluate owing to multiple reactions or more than one target isotope.

TABLE 4—*Concluded*

ISOTOPE	HALF-LIFE	RADIATIONS	REACTION	YIELD IN MICROCURIES PER MICROAMPERE HOUR				ISOTOPE RATIO
				8-M.e.v. deuterons	14-M.e.v. deuterons	19-M.e.v. deuterons	38-M.e.v. α-particles	
$_{50}Sn^{113}$..	105 days	K, e^-, γ	Cd(α,xn)				(0.50)	†
$_{51}Sb^{110}$..	6.0 days	K, e^-, γ	Sn(d,xn)			(2)		†
$_{51}Sb^{122}$..	2.8 days	β^-, e^-, γ						
$_{52}Te^{121m}$.	143 days	e^-, γ	Sb^{121}(d, $2n$)		(0.16)			1.8
$_{53}I^{131}$....	8.0 days	β^-, e^-, γ	Te^{130}(d,p) Te^{131}(d,n)	1.0	20.0			†
$_{56}Ba^{133m}$.	38.8 hr.	I.T., e^-, γ	Cs^{133}(d, $2n$)		(100)			†
$_{74}W^{181}$...	140 days	K, e^-, γ	Ta^{181}(d, $2n$)			(0.005)		†
$_{75}Re^{183}$..	240 days	K, e^-, γ	Ta(α,xn)				(0.05)	†
$_{75}Re^{184}$..	52 days	K, e^-, γ						
$_{77}Ir^{190}$..	10.7 days	K, e^-, γ	Os(d,xn)			(2)		†
$_{77}Ir^{192}$..	70 days	K, e^-, γ						
$_{82}Pb^{203}$..	52 hr.	K, e^-, γ	Tl^{203}(d, $2n$)			(1.5)		3.4
$_{83}Bi^{206}$..	6.4 days	K, e^-, γ	Pb^{206}(d, $2n$)			(10)		3.9
$_{84}Po^{210}$..	138 days	α, γ	Bi^{209}(d,n)		2.0			1
$_{84}Po^{210}$..	138 days	α, γ	Pb^{208}(α, $2n$)				0.85	2
$_{85}At^{211}$..	7.5 hr.	α, K	Bi^{209}(α, $2n$)				(100)‡	1

‡ Yield for 29-M.e.v. helium ions.

III. General Considerations in Preparation of Carrier-Free Radioisotopes

A. 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*) coprecipitation, (*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 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 solutions of sodium chloride, sodium sulfate, or other nontoxic salts. Second, because these specifications are generally high enough to meet the requirements of almost all other types of investigation requiring carrier-free radioisotopes.

1. Coprecipitation

The separation of submicrogram quantities of radioelements from solution by means of nonisotopic precipitates usually involves a subsequent separation of the radioelement from the carrier precipitate. It is desirable, therefore, to use, as coprecipitating agents, substances which can be easily separated with a minimum amount of chemical and physical manipulation. True coprecipitation processes (51, 61) (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 coprecipitation processes. This factor has, however, become somewhat less limiting with the development of the ion-exchange technique (127).

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 the true coprecipitation reaction, they have the important advantage 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 5 is a résumé of some important coprecipitation-scavenging types of precipitation reactions. A serious disadvantage of coprecipitation 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, coprecipitation reactions, particularly those involving adsorption phenomena, are frequently specific only under rigidly controlled experimental conditions (61).

2. 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

TABLE 5

Classification of separation processes used in the isolation of carrier-free radioisotopes

References to the detailed methods outlined in table 6 are given in parentheses

RADIOISOTOPE	PRODUCED BY	SEPARATION PROCESSES WHICH HAVE BEEN EMPLOYED						
		Coprecipitation	Solvent extraction	Ion exchange	Gas evolution, vaporization, and distillation	Radiocolloid	Leaching	Electrodeposition
H^3	Be(d,2α) Li(n,α)				(66, 97)			
Be^7	Li(d,n)	(24, 65)	(2, 24, 65)			(54)		
C^{11}	B(d,n)				(12, 109)			
C^{14}	N(n,p)				(96, 108, 134)			
N^{13}	C(d,n)				(95, 105)			
F^{18}	O(α,pn)						(52)	
$Na^{22, 24}$	Mg(d,α)			(5)			(27, 48, 63, 118)	
Mg^{27}	Al(d,2p)					(53)		
P^{32}	S(n,p)	(16, 114)	(30, 32)	(16)				(47)
S^{35}	Cl(d,α) Cl(n,p)			 (110)			(68, 114, 131)	
A^{37}	Cl(d,2n)				(52)			
$K^{42, 43}$	A(α,pn)						(99)	
Ca^{45}	Sc(n,p) Sc(d,2p)	(72)	(9)				(72)	
Sc^{46}	Ti(d,α)					(39)		
V^{48}	Ti(d,2n)						(58)	
Cr^{51}	V(d,2n)	(43)						
$Mn^{52, 54}$	Fe(d,α)	(89)	(89)					

TABLE 5—*Continued*

RADIOISOTOPE	PRODUCED BY	SEPARATION PROCESSES WHICH HAVE BEEN EMPLOYED						
		Coprecipitation	Solvent extraction	Ion exchange	Gas evolution, vaporization, and distillation	Radiocolloid	Leaching	Electrodeposition
$Mn^{52, 54}$	Cr(d,n)	(55)	(55)					
Fe^{59}	Co(d,2p) Co(n,p)		(70)					
$Co^{56, 57, 58}$	Fe(d,xn)						(89)	
$Cu^{64, 67}$	Zn(d,2p) Zn(n,p)	(4)	(59, 113)					(31)
Zn^{65}	Cu(d,2n)		(120)				(120)	
Ga^{67}	Zn(d,xn)		(49)					
Ge^{71}	Ga(d,2n)				(82)			
As^{74}	Ge(d,2n)				(81, 83)			
Se^{75}	As(d,2n)	(37)			(37)			
$Br^{80, 82}$	Se(d,2n)				(36)			
Kr^{79}	Br(d,2n)				(10, 52)			
Sr^{85}	Rb(d,2n)	(98)	(98)					
$Sr^{89, 90}$	U(n,f)			(7, 127)				
Y^{88}	Sr(d,2n)					(78)		
Y^{91}	U(n,f)			(71, 127)				
Zr^{89}	Y(d,2n)	(98)						
Zr^{95}	U(n,f)	(18, 98)	(18)	(127)				
Cb^{95}	U(n,f)	(18, 98)	(18)					
Mo^{99}	Zr(α,n)		(121)					
Tc^{95}	Mo(d,n)				(6, 90)			
Tc^{99}	U(n,f)	(101)			(101)			
Ru^{103}	U(n,f)	(98)			(98)			

TABLE 5—*Continued*

RADIOISOTOPE	PRODUCED BY	SEPARATION PROCESSES WHICH HAVE BEEN EMPLOYED						
		Coprecipitation	Solvent extraction	Ion exchange	Gas evolution, vaporization, and distillation	Radiocolloid	Leaching	Electrodeposition
Pd^{103}	Rh(d,2n)	(44)			(44)			
$Ag^{105, 106, 111}$	Pd(d,2n)	(57)						
Ag^{111}	Pd^{111} β-decay							(49)
Cd^{109}	Ag(d,2n)	(92)						
$In^{111, 114}$	Cd(α,pn)	(91)						
Sn^{113}	Cd(α,xn)				(121)			
$Sb^{120, 122}$	Sn(d,xn)	(91)			(91)			
$Te^{127, 129}$	U(n,f)	(98)			(98)			
I^{131}	Te(d,xn)	(98)			(98, 102)			
I^{131}	U(n,f)				(1, 110)			
I^{131}	Te^{131} β-decay				(1, 84, 110)			
Xe^{127}	I(d,2n)				(52, 98)			
Cs^{131}	Ba^{131} β-decay						(100)	
Cs^{137}	U(n,f)	(45, 98)		(100)				
Ba^{133}	Ca(d,2n)						(79, 98)	
Ba^{140}	U(n,f)			(112, 127)				
La^{140}	Ba^{140} β-decay	(98)	(98)	(112, 137)				
Rare earths: Z = 58 to Z = 63	U(n,f)			(100, 110, 127)				

TABLE 5—*Concluded*

RADIOISOTOPE	PRODUCED BY	SEPARATION PROCESSES WHICH HAVE BEEN EMPLOYED						
		Coprecipitation	Solvent Extraction	Ion exchange	Gas evolution, vaporization, and distillation	Radiocolloid	Leaching	Electrodeposition
Rare earths: $Z = 64$ to $Z = 71$.....	$Z(p,xn)$ (d,xn) (α,xn)	(132)		(132)				
$Ta^{177, 178, 180}$...	$Hf(d,xn)$	(56)		(56)				
$Re^{183, 184}$.....	$Ta(\alpha,xn)$	(41, 43)			(41, 43)			
Os^{185}.....	$W(\alpha,xn)$		(42)					
$Bi^{204, 206}$.....	$Pb(d,2n)$					(40)		
Po^{210}.....	$Pb(\alpha,xn)$	(93)	(93)					
Po^{210}.....	$Bi(d,n)$	(93)	(93)					
$At^{210, 211}$.....	$Bi(\alpha,xn)$				(35, 67)			

procedure in a few cases. These are described in table 5. 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.

3. 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 phenomenon is not entirely clear, it has been suggested (51, 78) that the radioelement becomes adsorbed on colloidal impurities which are normally present in the solution. Adsorption may also occur on the walls of the containing vessel. This phenomenon 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 sepa-

rated by use of this technique are shown in table 5. 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.

4. 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, and (*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 5. 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 the electrode material, necessitating a subsequent separation to obtain a chemically pure solution of the activity suitable for use in biological systems.

5. Ion exchange

The separation of trace amounts of radioelements by selective elution from ion-exchange columns has become an increasingly 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 complex-forming 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 (127). 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 complex-forming 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 thorough washing with water, the carrier-free radioisotopes are selectively eluted with 5 per cent 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 neigh-

boring 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 complex-forming 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 N 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 5.

6. Solvent extraction

The selective extraction of a radioactive tracer as a nonpolar compound or complex from an aqueous solution by an immiscible organic solvent is frequently the most satisfactory method of separating the activity from nonisotopic 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 nonisotopic 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 coprecipitation.

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 submicro amounts of the substance will be extractable. For example, carrier-free radioiron cannot be extracted into ethyl ether from 6 N hydrochloric acid, 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 the 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 nonpolar complexes or chelates with carrier-free radioelements. Compounds which form soluble nonpolar complexes whose dissociation constants are pH dependent are particularly useful in separating a carrier-free radioelement from a mixture. Extraction separations are summarized in table 5.

7. Distillation (volatilization)

Carrier-free radioelements which have been separated from solution, melts, and solids by volatilization or distillation processes are shown in table 5. 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 submacrogram 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 (52).

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.

B. PURITY CONSIDERATIONS

1. 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 (111) 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 forms of high purity have been discussed elsewhere (23).

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 by use of 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 weighable 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.

TABLE 6

Outline of methods for the production and isolation of carrier-free radioisotopes

RADIOISOTOPE	PRODUCED BY	DESCRIPTION OF SEPARATION PROCESSES
$_1H^3$.....	Be(d,t) Li(n,α)	The H^3 is released by heating the Be in a stream of oxygen or LiF in an evacuated quartz tube. The H^3 is condensed as H_2^3O after passage over hot CuO (66, 97).
$_4Be^7$.....	Li(d,n)	Be^7 is separated from Li by coprecipitation on $Fe(OH)_3$ from dilute NH_4OH solution. The $Fe(OH)_3$ containing the Be^7 is dissolved in 6 *N* HCl and Fe is extracted with ether (24, 65). The Li is dissolved in water. 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 (54). Be^7 is extracted from aqueous solution at pH 5–6 by thenoyl trifluoroacetone in benzene. The activity is back-extracted with concentrated HCl (2).
$_6C^{11}$.....	B(d,n)	$C^{11}O$ and $C^{11}O_2$ are expelled from a B_2O_3 target during bombardment. The $C^{11}O$ and $C^{11}O_2$ are passed over hot CuO, and the $C^{11}O_2$ is condensed in a liquid-air trap (12, 109).
$_6C^{14}$.....	N(n,p)	The $C^{14}O$ and $C^{14}O_2$ formed during bombardment of an 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 (96, 108, 134). $Ca(NO_3)_2$ 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 (110).
$_7N^{13}$.....	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 (95, 105).
$_9F^{18}$.....	O($d,2n$)	Liquid H_2O may be bombarded in an adequately cooled thin-window target to produce an aqueous solution of F^{18} directly (101, 128).
$_{11}Na^{22,\ 24}$.....	Mg(d,α)	The $Na^{22,\ 24}$ is dissolved out of MgO, $Mg(OH)_2$, or $MgCO_3$ almost quantitatively with water (27, 48, 118). $Na^{22,\ 24}$ does not coprecipitate with $Mg(NH_4)_2(CO_3)_2 \cdot 4H_2O$ (63). A separation of Na^{22} from weighable amounts of Mg may be obtained by the use of ion-exchange techniques (5).

TABLE 6—*Continued*

RADIOISOTOPE	PRODUCED BY	DESCRIPTION OF SEPARATION PROCESSES
$_{12}Mg^{27}$.....	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 (53).
$_{15}P^{32}$.....	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 (16, 114). 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 (30, 32). P^{32} is removed from irradiated CS_2 by the use of electrodes immersed in the liquid (47).
$_{16}S^{35}$.....	Cl(d,α) Cl(n,p)	KCl, NaCl, and $FeCl_3$ are used as target materials. Separation of S^{35} from irradiated KCl or NaCl is effected by taking advantage of the insolubility of these salts in concentrated 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 (68, 114, 131). 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 an ion-exchange column (110).
$_{18}A^{37}$.....	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* (52).
$_{19}K^{42,\ 43}$.....	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 (99).
$_{20}Ca^{45}$.....	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 coprecipitated on $BaCO_3$. Separation from the Ba is effected by taking advantage of the insolubility of $BaCl_2$ in concentrated HCl–ether solution (72).

TABLE 6—*Continued*

RADIOISOTOPE	PRODUCED BY	DESCRIPTION OF SEPARATION PROCESSES
		The Sc_2O_3 is dissolved in 12 *N* 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 reëxtracted with 0.5 *M* thenoyl trifluoroacetone. The Ca^{45} in the benzene fraction is back-extracted with H_2O or dilute HCl (9).
$_{21}Sc^{44, 46, 47, 48}$...	Ti(d,α) Ti($d,2p$)	The Ti metal target is dissolved in a minimum volume of H_2SO_4. The solution is slowly added to excess 8 *N* NH_4OH containing 30 per cent H_2O_2 to give a clear "solution" of the soluble pertitanate containing the Sc activity as radiocolloidal aggregates. This solution is then drawn through filter paper, which retains over 95 per cent of the Sc activity as adsorbed radiocolloid, which is removed by treatment with dilute HCl (39).
$_{23}V^{48}$.....	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 (58).
$_{24}Cr^{51}$.....	V($d,2n$) V(p,n)	The V target is dissolved in HNO_3. The solution is diluted to 6 *N* 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 (43).
$_{25}Mn^{52, 54}$.....	Fe(d,α)	The Fe target is dissolved in HCl. All but a few milligrams of Fe is extracted with ether. The $Mn^{52, 54}$ in the tetrapositive state is quantitatively coprecipitated 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 (89).
	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 coprecipitated on the $Fe(OH)_3$, which is dissolved in 6 *N* HCl and extracted with ether to remove Fe carrier (55).
$_{26}Fe^{59}$.....	Co($d,2p$) Co(n,p)	The Co is dissolved in 3 *N* HNO_3 and the pH of the solution is adjusted to 4.0–7.0 by the addition of NH_4OH–$NH_4C_2H_3O_2$.

TABLE 6—*Continued*

RADIOISOTOPE	PRODUCED BY	DESCRIPTION OF SEPARATION PROCESSES
		1 ml. of a saturated aqueous solution of acetylacetone 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 $HClO_4$ (70).
	Co(d,2p)	The Co target is dissolved in HCl. Excess NH_4OH is added and the resultant clear "solution" is passed through two consecutive Whatman No. 50 filter papers, which quantitatively removed the Fe^{59} as adsorbed radiocolloid. The activity is removed from the filter paper with dilute HCl (43).
$_{27}Co^{55, 56, 57, 58}$	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. Ammonium salts are destroyed with HNO_3 (89).
$_{29}Cu^{64, 67}$	Zn(d,2p)	The Zn target is dissolved in HCl, diluted to 5.5 N, and the 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 0.001 per cent dithizone. The CCl_4 phase is evaporated to dryness and organic material is removed by heating to 500°C. (59, 113).
	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 (4). $ZnCl_2$ target is dissolved in water. Cu^{64} is concentrated by addition of a small amount of Zn, which is then dissolved in acid and Cu^{64} is deposited on polished Pt foil from 0.05 N H_2SO_4 saturated with H_2 (31).
$_{30}Zn^{65}$	Cu(d,2n)	Cu target is dissolved in HNO_3. Excess HCl is added to destroy HNO_3 and the solution is neutralized to 0.25 N HCl with NH_4OH. CuS is precipitated with H_2S. Zn^{65} is not coprecipitated. The supernatant is adjusted to pH 5–5.5 with $NaC_2H_3O_2$ 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 0.02 N HCl (120).
$_{31}Ga^{67}$	Zn(d,n) Zn(d,2n)	Zn target is dissolved in HCl and adjusted to 6 N. Ga^{67} is extracted with diethyl ether (49).
$_{32}Ge^{71}$	Ga(d,2n)	The Ga target is dissolved in 48 per cent HBr and the solution is distilled. The Ge^{71} is obtained in the first few milliliters of

TABLE 6—*Continued*

RADIOISOTOPE	PRODUCED BY	DESCRIPTION OF SEPARATION PROCESSES
		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 (82).
$_{33}As^{74}$	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 nonvolatile 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 (81, 83).
$_{34}Se^{75}$	As(d,2n)	The As is dissolved in aqua regia. HCl is added to destroy excess HNO_3. The solution is adjusted to 3 N. 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 (37).
$_{35}Br^{80,\ 82}$	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 and does not extract the CCl_4 (36).
$_{36}Kr^{79}$	Br(d,2n)	The Kr^{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* (10, 52).
$_{38}Sr^{85}$	Rb(d, 2n)	The RbCl is dissolved in 0.1 N 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 6 N HCl and the Fe is extracted with ether (98).
$_{38}Sr^{89,\ 90}$	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 per cent citrate at pH 5 (7, 127). (A more detailed description of fission-product separation by ion-exchange methods is given in the section on rare earths.)
$_{39}Y^{88}$	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 (78).
$_{39}Y^{91}$	U(n,f)	Y^{91} is separated from rare-earth fission products by complex elution from Amberlite IR-1 resin with 5 per cent citrate at pH 2.7 (71, 127). (A more detailed description of fission-product separations is given in the section on rare earths.)

TABLE 6—*Continued*

RADIOISOTOPE	PRODUCED BY	DESCRIPTION OF SEPARATION PROCESSES
$_{40}Zr^{89}$	$Y(d,2n)$	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 milliliters 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 6 *N* HCl and Fe is extracted with ether (98).
$_{40}Zr^{95}$	$U(n,f)$	Oxalic acid is used as a specific complex-forming agent to remove Zr^{95} and Cb^{95} from mixed fission products on Amberlite IR-1 columns. 0.5 per cent oxalic acid strips Zr^{95}–Cb^{95} quantitatively; none of the trivalent or divalent elements is removed by this procedure (127). Zr^{95} is separated from Cb^{95} daughter by (*a*) coprecipitation of Cb^{95} on MnO_2 from 10 *N* HNO_3 and (*b*) extraction of Zr^{95} from HNO_3 or $HClO_4$ using thenoyl trifluoroacetone (18). The bulk of the uranyl nitrate target material is removed by ether extraction. To the concentrated solution of fission products are added a few milligrams of $FeCl_3$, hydroxylamine, and excess NH_4OH. Zr^{95} and other insoluble fission products, in the presence of NH_4OH, are coprecipitated 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 (98).
$_{41}Cb^{95}$	$U(n,f)$	See Zr^{95} procedures.
$_{42}Mo^{99}$	$Zr(\alpha,n)$	The ZrO_2 target is dissolved in the presence of Cb "holdback" carrier by boiling in 12 *N* H_2SO_4 down to fuming. NaCl and HCl are added, following cooling and dilution with water, to bring the (H^+) and (Cl^-) to 6 *N*. The Mo is extracted with ether saturated with HCl (121).
$_{43}Tc^{95}$	$Mo(d,n)$	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 (6, 90).
$_{43}Tc^{99}$	$U(n,f)$	The U metal is dissolved in 37 per cent HCl and the UCl is oxidized to UO_2Cl_2 with H_2O_2 and Br_2 at 60°C. $PtCl_4$ is added and the solution is saturated with H_2S at 90°C. The Tc^{99} is coprecipitated 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 evaporated to incipient dryness. 18 *N* H_2SO_4 is added and the Tc^{99} is distilled (101).

TABLE 6—*Continued*

RADIOISOTOPE	PRODUCED BY	DESCRIPTION OF SEPARATION PROCESSES
$_{44}Ru^{97, 103}$.....	Mo(α, xn)	The Mo target is fused with KOH and KNO_3. The resultant melt is extracted with water. The basic solution is transferred to a distilling flask and the carrier-free $Ru^{97, 103}$ is volatilized by passing Cl_2 through the solution with heating. The Ru activity is collected in a trap filled with 12 *N* HCl. The distillate is evaporated to a small volume, treated with excess KOH, and the $Ru^{97, 103}$ is redistilled as above (43).
$_{44}Ru^{103}$.....	U(n,f)	The U metal is dissolved in HNO_3 and adjusted to 3 *N*. 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 3 *N* HCl. The distillate is made strongly alkaline and Ru^{103} is distilled from alkaline solution after oxidation with Cl_2 (98).
$_{45}Rh^{101, 102}$.....	Ru(d, xn)	The Ru target is fused with Na_2O_2 and the melt is digested with HCl. After centrifugation, the acid solution is made basic with KOH and the Ru is volatilized by passing Cl_2 through the solution with heating. The residue is acidified, 5 mg. of Fe^{+++} is added, and the $Rh^{101, 102}$ is coprecipitated on $Fe(OH)_3$ with the addition of KOH. The $Fe(OH)_3$ is dissolved in 6 *N* HCl and Fe is separated by ether extraction (43).
$_{46}Pd^{103}$.....	Rh(d, $2n$)	The Rh is fused with $KHSO_4$ and the fused mass is dissolved in water. HCl is added together with milligram 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 $HClO_4$ (44).
$_{47}Ag^{105, 106, 111}$...	Pd(d,n)	Ag^{105} is separated by coprecipitation on Hg_2Cl_2, which is removed by volatilization (57).
$_{47}Ag^{111}$.....	Pd^{111}	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. (50).
$_{48}Cd^{109}$.....	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 combined in a complex with NH_4CNS. The solution is adjusted to pH 5 with sodium acetate, and the Cd^{109} is extracted with chloroform containing 5 per cent pyridine (92).
$_{49}In^{111, 114}$.....	Cd(α,p) Cd(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 in the In^{114} quantitatively in the presence of the NH_4OH. Fe is separated by ether extraction (91).
$_{50}Sn^{113}$.....	Cd(α,n) Cd(α, $2n$)	The Cd is dissolved in HNO_3 and the solution is evaporated to incipient dryness, diluted with H_2O, and made alkaline with NH_4OH after the addition of $FeCl_3$. The Sn^{113} and In^{114}

TABLE 6—*Continued*

RADIOISOTOPE	PRODUCED BY	DESCRIPTION OF SEPARATION PROCESSES
		are carried on the $Fe(OH)_3$ precipitate. The $Fe(OH)_3$ is dissolved in a minimum volume of 36 *N* 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 (91).
$_{51}Sb^{120, 122}$.....	$Sn(d,n)$ $Sn(d,2n)$	The Sn target is dissolved in aqua regia. HCl is added to destroy excess HNO_3 and the solution is adjusted to 0.1 *N* 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 (91).
$_{52}Te^{127, 129}$.....	$U(n,f)$	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 5 *N* in HF. The $Te^{127, 129}$ is then coprecipitated on RuS_2. The RuS_2 is decomposed with HNO_3 and the residue is boiled with $HClO_4$ to expel the Ru (98).
$_{53}I^{131}$.....	$Te(d,n)$ $Te(d,2n)$	The Te is dissolved in CrO_3–H_2SO_4 solution. Oxalic acid is added and the I* is distilled into NaOH (102).
	$U(n,f)$	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 per cent 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 NaOH–Na_2SO_3 solution (1, 110).
	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, followed by solid oxalic acid and the I^{131} is distilled into dilute Na_2SO_3–Na_2CO_3 solution (84). Te target is dissolved in 18 *N* H_2SO_4–50 per cent CrO_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 (1, 110).
$_{54}Xe^{127}$.....	$I(d,2n)$	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* (52, 98).

* 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 (98).

TABLE 6—*Continued*

RADIOISOTOPE	PRODUCED BY	DESCRIPTION OF SEPARATION PROCESSES
$_{55}Cs^{131}$	Ba^{131} β^--decay	$BaCl_2$ is precipitated from 12 *N* HCl saturated with Cl_2 at 0°C. Cs^{131} remains in the supernatant. Further purification is obtained by scavenging with $La(OH)_3$ (34).
$_{55}Cs^{137}$	U(*n*,*f*)	Cs^{137} is separated from the divalent or trivalent fission product ions by preferential elution from Amberlite IR-1, using 0.25 *N* HCl (100). Cs^{137} is separated from fission mixture by coprecipitation on NH_4ClO_4 from $HClO_4$ solutions using absolute alcohol (45). Cs^{137} is separated from a fission-product mixture by coprecipitation on sodium cobaltinitrite and ammonium cobaltinitrite. The Co is removed as CoS from NH_4OH solution (98).
$_{56}Ba^{133}$	Cs(*d*,2*n*)	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 (79, 98).
$_{56}Ba^{140}$	U(*n*,*f*)	After the elution of the rare earths from Amberlite IR-1, using 5 per cent citrate at pH 2.7, the $Sr^{89, 90}$ and Ba^{140} are successively removed by 5 per cent citrate at pH 5 (112, 127).
$_{57}La^{140}$	Ba^{140} β^--decay Ba^{140} β^--decay	La^{140} is separated from Ba^{140}–La^{140} mixtures by elution from Amberlite IR-1, using 5 per cent citrate at pH 2.7 (112, 127). $BaCl_2$ is added to an HCl–Ba^{140} solution and precipitated by passing HCl gas through the solution at 0°C. The $BaCl_2$ containing the Ba^{140} is dissolved in H_2O. $FeCl_3$ carrier 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 6 *N* HCl (98).
57–63 La^{140} Pr^{143} Nd^{147} Pm^{147} Sm^{161} Eu^{155}	U(*n*,*f*)	A solution of all fission-produced species in 0.01–0.1 *N* HCl is passed through an Amberlite IR-1 resin column. All cations are adsorbed while the fission-produced anionic elements (Ru, Te, Tc) and any I present which pass through the acid are recovered in the effluent. Most of the Cs^{137} may be eluted at this point by the addition of 0.25 *N* HCl. The column is washed, and Cb^{95}–Zr^{95} is preferentially removed by 0.5 per cent 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 per cent citric acid at a pH of from 2.7 to 2.95. In practice a group of several elements may be eluted, then readsorbed and fractionated under more rigidly controlled conditions (100, 110, 127).

TABLE 6—*Continued*

RADIOISOTOPE	PRODUCED BY	DESCRIPTION OF SEPARATION PROCESSES
64–71 Gd Tb Dy Ho Er Tm Yb Lu	Z(p,xn) Z(d,xn) Z(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 2 N 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 eluted with 0.25 M citric acid adjusted to pH 3.05 with NH_4OH (93, 132).
$_{73}Ta^{177, 178, 179}$	Hf(d, xn)	The HfO_2 is dissolved in HNO_3–HF solution. HF is removed by evaporation with the addition of 16 N 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 (56).
$_{75}Re^{183, 184}$	Ta(α, xn)	The Ta metal target is dissolved in a minimum volume of 16 N HNO_3 containing 10 per cent 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 36 N H_2SO_4. The $Re^{183, 184}$ is distilled at 240°C. with the addition of 9 N HBr and coprecipitated on CuS after the removal of HBr and HNO_3. Separation from the Cu is effected by precipitating CuO from alkaline solution (41, 43).
	W(d,xn)	The W target is fused with KOH–KNO_3 and the fused mass is extracted with water. The solution is acidified with 16 N HNO_3, precipitating tungstic acid which is removed by centrifugation. The carrier-free $Re^{183, 184}$ is isolated from the HNO_3 supernatant, using the distillation procedure described above (42).
$_{76}Os^{185}$	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 5 N HNO_3 and collected in a trap filled with 5 N HNO_3. Extraction of the HNO_3 distillate with ether removes the Os^{185}, which is back-extracted with NaOH (42).
$_{77}Ir^{190, 192}$	Os(d,xn)	The Os powder is dissolved in aqua regia and volatilized with the addition of excess HNO_3. The residue is fumed with H_2SO_4 to remove Re activity and then heated to dryness. The carrier-free $Ir^{190, 192}$ is redissolved in 12 N HCl and the resultant solution is evaporated to dryness on NaCl (56).

TABLE 6—*Concluded*

RADIOISOTOPE	PRODUCED BY	DESCRIPTION OF SEPARATION PROCESSES
$_{82}Pb^{203}$.....	Tl(d,2n)	The Tl_2O_3 target material is dissolved in dilute HNO_3. Tl^{+++} is reduced to Tl^+ with SO_2. Five milligrams of Fe^{+++} is added and the solution is made basic with NH_4OH. The Pb^{203} is coprecipitated with the $Fe(OH)_3$ and the Fe removed by ether extraction (56).
$_{83}Bi^{204, 206}$.....	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 per cent NaOH. This "solution" is drawn through filter paper which retains over 98 per cent of the Bi activity as adsorbed radiocolloid. Dilute HCl removes the $Bi^{204, 206}$ quantitatively (40).
$_{84}Po^{210}$.....	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 3 N and Te carrier is precipitated with SO_2, removing At and Po. The Te is reprecipitated from 6 N HCl. Under these conditions At is coprecipitated and Po remains in the supernatant. Po in the HCl solution is extracted with an equal volume of 20 per cent tributyl phosphate in diethyl ether. The activity is back-extracted with HNO_3 (93).
$_{85}At^{210, 211}$.....	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 (35, 67).

2. Radiochemical contamination

Although many of the possible sources of radiochemical contamination in radioisotope preparations can be eliminated by a proper choice of target materials, bombardment 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 the 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 (17).

C. 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 bom-

bardment 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 1 milliamp. 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-M.e.v. 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 pure metals or metallic alloys having a high melting point and a high heat conductivity. Even for these substances the power dissipation cannot usually 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 (87).

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 enclosure 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 alloys 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 very thin metal foils, usually of platinum or tantalum. Many different target designs have been developed for special uses (80, 104, 129, 133).

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IV. REFERENCES[3]

(1) BALLANTINE, D. S., AND COHN, W. E.: MDDC-1600 (June, 1947).
(2) BALOMEY, R. A., AND WISH, L.: AECD-2665 (August, 1949).
(3) BARSCHALL, H. H., AND BATTAT, M. E.: Phys. Rev. **70,** 245 (1946).
(4) BORN, H. J., AND DREHMAN, U.: Naturwissenschaften **32,** 159 (1945).
(5) BOUCHEZ, R., AND KAYAS, G.: Compt. rend. **228,** 1222 (1949).
(6) BOYD, G. E., LARSON, O. V., AND MOLTA, M. E.: AECD-2151 (June, 1948).
(7) BOYD, G. E., SCHUBERT, J., AND ADAMSON, A. W.: MDDC-708 (1947).
(8) BRADT, H. L., AND TENDAM, D. J.: Phys. Rev. **72,** 1117 (1947).
(9) BROIDO, A.: AECD-2616 (June, 1949).
(10) BROWN, S. C., IRVINE, J. W., AND LIVINGSTON, M. S.: J. Chem. Phys. **12,** 132 (1944).
(11) BUCK, J. H.: Phys. Rev. **54,** 1025 (1938).
(12) CAILLAT, R., AND SUE, P.: Bull. soc. chim. France **1949,** 687.
(13) CLARKE, E. T.: Phys. Rev. **71,** 187 (1947).
(14) CLARKE, E. T., AND IRVINE, J. W., JR.: Phys. Rev. **66,** 231 (1944).
(15) CLARKE, E. T., AND IRVINE, J. W., JR.: Phys. Rev. **69,** 680 (1946).
(16) COHN, W. E.: Isotope Branch Circular No. C-1, U. S. Atomic Energy Commission (1947).
(17) COHN, W. E.: Anal. Chem. **20,** 498 (1948).
(18) CONNICK, R. E., AND MCVEY, W. H.: J. Am. Chem. Soc. **71,** 3182 (1949).
(19) CORK, J. M.: Phys. Rev. **70,** 563 (1946).
(20) CORK, J. M., AND CURTIS, R. B.: Phys. Rev. **55,** 1264 (1939).
(21) CORK, J. M., AND HALPERN, J.: Phys. Rev. **57,** 667 (1940).
(22) CORK, J. M., HALPERN, J., AND TATEL, H.: Phys. Rev. **57,** 371 (1940).
(23) COVEY, E. H.: UCRL-266 (March, 1949).
(24) CROWLEY, J., HAMILTON, J. G., AND SCOTT, K.: J. Biol. Chem. **177,** 975 (1949).
(25) CUNNINGHAM, B. B., HOPKINS, H. H., LINDER, M., MILLER, D. R., O'CONNOR, P. R., PERLMAN, I., SEABORG, G. T., AND THOMPSON, R. C.: Phys. Rev. **72,** 739 (1947).
(26) DELSASSO, L. A., RIDENOUR, L. N., SHERR, R., AND WHITE, M. G.: Phys. Rev. **55,** 113 (1939).
(27) DREHMANN, V.: Naturwissenschaften **33,** 24 (1946).
(28) DU BRIDGE, L. A., BARNES, S. W., BUCK, J. H., AND STRAIN, C. V.: Phys. Rev. **53,** 447 (1938).
(29) EDWARDS, R. R., AND DAVIES, T. H.: Nucleonics **2,** 44 (1948).
(30) ERBACHER, O.: Z. physik. Chem. **B42,** 173 (1939).
(31) ERBACHER, O., HERR, W., AND EGIDI, V.: Z. anorg. Chem. **256,** 41 (1948).
(32) ERBACHER, O., AND PHILIPP, K.: Ber. **69,** 893 (1936).
(33) FINKLE, B., AND COHN, W. E.: AECD-2556-F (March, 1949).
(34) FAJANS, K., AND VOIGT, A. F.: Phys. Rev. **60,** 619 (1941).
(35) GARRISON, W. M., GILE, J. D., MAXWELL, R. D., AND HAMILTON, J. G.: UCRL-741 (May, 1950).
(36) GARRISON, W. M., AND HAMILTON, J. G.: Unpublished data.
(37) GARRISON, W. M., MAXWELL, R. D., AND HAMILTON, J. G.: J. Chem. Phys. **18,** 155 (1950).
(38) GHOSHAL, S. N.: Phys. Rev. **73,** 417 (1948).
(39) GILE, J. D., GARRISON, W. M., AND HAMILTON, J. G.: UCRL-921 (September, 1950).

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[3] Those references designated by the symbols AECD and MDDC refer to declassified U. S. Atomic Energy Commission publications. The symbol UCRL refers to a University of California Radiation Laboratory publication.

(40) GILE, J. D., GARRISON, W. M., AND HAMILTON, J. G.: UCRL-1017 (December, 1950).
(41) GILE, J. D., GARRISON, W. M., AND HAMILTON, J. G.: J. Chem. Phys. **18,** 995 (1950).
(42) GILE, J. D., GARRISON, W. M., AND HAMILTON, J. G.: J. Chem. Phys. **18,** 1419 (1950).
(43) GILE, J. D., GARRISON, W. M., AND HAMILTON, J. G.: To be published.
(44) GILE, J. D., HAYMOND, H. R., GARRISON, W. M., AND HAMILTON, J. G.: To be published.
(45) GLENDENIN, L. E., AND NELSON, C. M.: AECD-2556-E (March, 1949).
(46) GOECKERMANN, R. H., AND PERLMAN, I.: Phys. Rev. **73,** 1127 (1948).
(47) GOVAERTS, J.: J. Chem. Phys. **36,** 130 (1939).
(48) GOVAERTS, J.: Bull. soc. roy. sci. Liége **9,** 38 (1940).
(49) GRAHAME, D. C., AND SEABORG, G. T.: Phys. Rev. **54,** 240 (1938).
(50) GRIESS, J. C., JR., AND ROGERS, L. B.: AECD-2299 (September, 1948).
(51) HAHN, O.: *Applied Radiochemistry.* Cornell University Press, Ithaca, New York (1936).
(52) HAMILTON, J. G.: Unpublished data (1950).
HAMILTON, J. G.: Unpublished data (1942).
HAMILTON, J. G.: Unpublished data (1942).
(53) HAYMOND, H. R., BOWERS, J. Z., GARRISON, W. M., AND HAMILTON, J. G.: J. Chem. Phys. **18,** 1119 (1950).
(54) HAYMOND, H. R., GARRISON, W. M., AND HAMILTON, J. G.: UCRL-920 (September, 1950).
(55) HAYMOND, H. R., GARRISON, W. M., AND HAMILTON, J. G.: UCRL-1033 (December, 1950).
(56) HAYMOND, H. R., GARRISON, W. M., AND HAMILTON, J. G.: To be published.
(57) HAYMOND, H. R., LARSON, K. H., MAXWELL, R. D., GARRISON, W. M., AND HAMILTON, J. G.: J. Chem. Phys. **18,** 391 (1950).
(58) HAYMOND, H. R., MAXWELL, R. D., GARRISON, W. M., AND HAMILTON, J. G.: J. Chem. Phys. **18,** 756 (1950).
(59) HAYMOND, H. R., MAXWELL, R. D., GARRISON, W. M., AND HAMILTON, J. G.: J. Chem. Phys. **18,** 901 (1950).
(60) HENDERSON, M. C.: Phys. Rev. **48,** 855 (1935).
(61) HEVESY, G., AND PANETH, F. H.: *Radioactivity.* Oxford University Press, London (1938).
(62) HURST, D. G., LATHAM, R., AND LEWIS, W. B.: Proc. Roy. Soc. (London) **A174,** 126 (1940).
(63) IRVINE, J. W., JR., AND CLARKE, E. T.: J. Chem. Phys. **16,** 686 (1948).
(64) Isotopes Division, Atomic Energy Commission, Oak Ridge, Tennessee, Catalogue No. 3 (July, 1949).
(65) JACOBSON, L., AND OVERSTREET, R.: Unpublished data (1943).
(66) JACOBSON, L., OVERSTREET, R., AND HAMILTON, J. G.: Unpublished data (1943).
(67) JOHNSON, G. L., LEININGER, R. F., AND SEGRÈ, E.: J. Chem. Phys. **17,** 1 (1949).
(68) KAMEN, M. D.: Phys. Rev. **60,** 537 (1941).
(69) KELLEY, E. L., AND SEGRÈ, E.: Phys. Rev. **75,** 999 (1949).
(70) KENNY, A. W., MATON, W. R. E., AND SPRAGG, W. T.: Nature **165,** 483 (1950).
(71) KETTLE, B. H., AND BOYD, G. E.: J. Am. Chem. Soc. **69,** 2800 (1947).
(72) KHYM, J. X., TOMPKINS, P. C., AND COHN, W. E.: MDDC-1214 (August, 1947).
(73) KLEMA, E. D., AND HANSON, A. O.: Phys. Rev. **73,** 106 (1948).
(74) KRAUS, J. D., AND CORK, J. M.: Phys. Rev. **52,** 763 (1937).
(75) KRISHNAN, R. S.: Proc. Cambridge Phil. Soc. **36,** 500 (1940).
(76) KRISHNAN, R. S.: Proc. Cambridge Phil. Soc. **37,** 186 (1941).
(77) KRISHNAN, R. S., AND NAHUN, E. A.: Proc. Soc. (London) **A180,** 333 (1942).
(78) KURBATOV, J. D., AND KURBATOV, M. H.: J. Phys. Chem. **46,** 441 (1942).

(79) KURBATOV, M. H., YU, FU-CHUN, AND KURBATOV, J. D.: J. Chem. Phys. **16,** 87 (1948).
(80) KURIE, F. N. D.: Rev. Sci. Instruments **10,** 199 (1939).
(81) LANZ, H., JR., AND HAMILTON, J. G.: MDDC-1596 (January, 1948).
(82) LANZ, H., JR., SCOTT, K. G., AND HAMILTON, J. G.: Unpublished data (1948).
(83) LANZ, H., JR., WALLACE, P. C., AND HAMILTON, J. G.: University of California Publications in Pharmacology **2,** 263 (1950).
(84) LEVY, M., KESTON, A. S., AND UDENFRIED, S.: J. Am. Chem. Soc. **70,** 2289 (1948).
(85) LIBBY, W. F.: J. Am. Chem. Soc. **69,** 2523 (1947).
(86) LINDNER, M., AND PERLMAN, I.: Phys. Rev. **78,** 499 (1950).
(87) LIVINGSTON, M. S.: J. Applied Phys. **15,** 128 (1944).
(88) LIVINGSTON, R. S., AND WRIGHT, B. T.: Phys. Rev. **58,** 656 (1940).
(89) MAXWELL, R. D., GILE, J. D., GARRISON, W. M., AND HAMILTON, J. G.: J. Chem. Phys. **17,** 1340 (1949).
(90) MAXWELL, R. D., AND HAYMOND, H. R.: Unpublished data (1948).
(91) MAXWELL, R. D., HAYMOND, H. R., GARRISON, W. M., AND HAMILTON, J. G.: J. Chem. Phys. **17,** 1005 (1949).
(92) MAXWELL, R. D., HAYMOND, H. R., GARRISON, W. M., AND HAMILTON, J. G.: J. Chem. Phys. **17,** 1006 (1949).
(93) MEINKE, W. N.: AECD-2738 (October, 1949).
(94) NEWSON, N.H.: Phys. Rev. **51,** 620 (1937).
(95) NORRIS, T. H., RUBEN, S., AND KAMEN, M. D.: J. Chem. Phys. **9,** 726 (1941).
(96) NORRIS, L. D., AND SNELL, A. H.: Science **105,** 265 (1947).
(97) O'NEAL, R. D., AND GOLDHABER, M.: Phys. Rev. **57,** 1086 (1940).
(98) OVERSTREET, R., JACOBSON, L., AND HAMILTON, J. G.: MDDC-1275 (1946).
(99) OVERSTREET, R., JACOBSON, L., AND STOUT, P. R.: Phys. Rev. **75,** 321 (1949).
(100) PARKER, G. W., AND LANTZ, P. M.: AECD-2160 (July, 1948).
(101) PARKER, G. W., REED, J., AND RUCH, J. W.: AECD-2043 (June, 1948).
(102) PERLMAN, I., MORTON, M. E., AND CHAIKOFF, I. L.: Endocrinology **30,** 487 (1942).
(103) Plutonium Project: J. Am. Chem. Soc. **68,** 2411 (1946).
(104) REID, A. F.: Rev. Sci. Instruments **18,** 501 (1947).
(105) RUBEN, S., HASSID, W. Z., AND KAMEN, M. D.: Science **91,** 578 (1940).
(106) RUBEN, S., AND KAMEN, M. D.: Phys. Rev. **59,** 349 (1941).
(107) RUBEN, S., AND KAMEN, M. D.: Phys. Rev. **60,** 537 (1941).
(108) RUBEN, S., AND KAMEN, M. D.: J. Applied Phys. **12,** 31 (1941).
(109) RUBEN, S., KAMEN, M. D., AND HASSID, W. Z.: J. Am. Chem. Soc. **62,** 3443 (1940).
(110) RUPP, A. F.: Private communication, April, 1950.
(111) SANDELL, E. B.: *Determination of Traces of Metals.* Interscience Publishers, Inc., New York (1944).
(112) SCHUBERT, J., AND RICHTER, J. W.: AECD-1986 (May, 1948).
(113) SCHULTZE, M. O., AND SIMMONS, S. J.: J. Biol. Chem. **142,** 97 (1942).
(114) Science **103,** 703 (1946).
(115) SEABORG, G. T.: Chem. Revs. **27,** 199 (1940).
(116) SEABORG, G. T., AND PERLMAN, I.: Rev. Modern Phys. **20,** 585 (1948).
(117) SEGRÈ, E., HALFORD, R. S., AND SEABORG, G. T.: Phys. Rev. **55,** 321 (1939).
(118) SEGRÈ, E., MACKENZIE, K. R., AND CORSON, D. R.: Phys. Rev. **57,** 1087 (1940).
(119) SEREN, L., FRIEDLANDER, H. N., AND TURKEL, S. H.: Phys. Rev. **72,** 888 (1947).
(120) STOUT, P. R.: Private communication.
(121) STOUT, P. R., AND MEAGHER, W. R.: Science **108,** 471 (1948).
(122) STRAIN, C. V.: Phys. Rev. **54,** 1021 (1938).
(123) SZILARD, L., AND CHALMERS, T. L.: Nature **134,** 462 (1934).
(124) TATEL, H. E., AND CORK, J. M.: Phys. Rev. **71,** 159 (1947).
(125) TEMMER, G. M.: Phys. Rev. **76,** 424 (1949).
(126) THORNTON, R. L.: Phys. Rev. **51,** 893 (1937).

(127) TOMPKINS, E. R., KHYM, J. X., AND COHN, W. E.: J. Am. Chem. Soc. **69,** 2769 (1947).
(128) VOLKER, J. E., HODGE, J. C., WILSON, H. J., AND VAN VOORHIS, S. N.: J. Biol. Chem. **134,** 535 (1940).
(129) WEIL, A. S., REID, A. F., AND DUNNING, J. R.: Rev. Sci. Instruments **18,** 556 (1947).
(130) WEISSKOPF, V. J., AND EWING, D. H.: Phys. Rev. **57,** 472 (1940).
(131) WILK, M. B.: Can. J. Research **27,** 475 (1949).
(132) WILKINSON, G. W., AND HICKS, H.: Phys. Rev. **75,** 1370 (1949).
(133) WILSON, R. R., AND KAMEN, M. D.: Phys. Rev. **54,** 1031 (1938).
(134) YANKWICH, R. E., ROLLEFSON, G. K., AND NORRIS, T. H.: J. Chem. Phys. **14, 131** (1940).