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RADIATION PROBLEMS ASSOCIATED WITH THE
HANDLING OF THE ACTINIDE ELEMENTS

by

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I. INTRODUCTION

The use of radioactive materials in research and industry has been prevalent for a considerable number of years, but the hazards associated with these materials have become recognized and appreciated only relatively recently. Thus, the recommended limits of exposure to gamma radiation have been gradually reduced as more and increasingly accurate information has become available on the effects of radiation on the human body.* While this increase in appreciation of the hazards has taken place, the number of workers in those fields utilizing radioactive materials has risen sharply. As a consequence of the latter, it has become desirable to provide information in readily intelligible terms to a broad spectrum of persons with widely varying degrees of training. Unfortunately, suitable written reviews and broad summaries on the hazards of handling radioactive materials are few.

In connection with the work of the Atomic Energy Commission, information on the hazards of handling radioactive materials has appeared scattered throughout the reported literature and in memoranda receiving little or no wide circulation. Only on a few occasions has an attempt been made to correlate such information into a coherent report. At this time considerable research and development work deals with the actinide elements, and therefore this review is designed to present an outline of the hazards likely to be encountered on handling these elements. Emphasis is directed toward problems appropriate to research and small-scale operations. Further, no particular attempt is made to cover the field completely, but it is hoped that this discussion, coupled with the attending references, will acquaint the reader with most of the problems and serve as a point of departure for those interested in more specific information.

The hazards of immediate interest are those arising from the radioactivity of the actinides themselves. Radioactivity from fission products which may be present in samples of the actinides is not considered appropriate to the present discussion. Chemical reactions and problems normally encountered in the control of reaction rates, fires, and the like are not unique to the actinide elements and are presumed to be general knowledge. The elements considered here, including thorium, protactinium, uranium, and the transuranic elements, are treated in varying degrees

*For a brief historical review, see Nuclear Safety 2, No. 3, p. 1 (1961).

of detail with emphasis on uranium, specifically uranium-233 and the chain of isotopes leading to its formation by neutron reaction, as well as plutonium. As is evident from this choice, emphasis is placed on elements and their isotopes which arise from the use of neutron reactors. Further, in each instance the type and magnitude of the hazard is stressed rather than the solution to certain problems. Finally, while a discussion of the hazards may well be useful in itself, certain nuclear data, such as modes of formation of the several isotopes, abundance, and specific activities, have been included to expand the usefulness of the review.

The hazards associated with the handling of radioactive materials are biological and are normally evaluated with reference to the human body. While reference is made to sources listing maximum permissible limits, the data developed for the individual nuclides are not presented in these terms. This lack of correlation is the result of the often great variation of the tolerated risk which may be found among the laboratories and production facilities engaged in work with actinide elements. The biological hazards are divided into two general classes: internal and external. The internal hazards, arising from the effects when the radioactive species are deposited within the body arise from two sources. The actinides, being heavy metals, act as chemical poisons similar to other nonradioactive heavy metals, such as lead. Although the chemical toxicity of the actinides is high, this aspect of their toxicity is generally not as important as the toxicity due to their radioactivity. For example, Langham⁽¹⁾ has pointed out that while, on the basis of its chemical toxicity, the maximum permissible body burden of plutonium is estimated to be about 40 mg, the high specific activity of plutonium limits the maximum permissible body burden to considerably less than that figure. Thus the most recent value for the maximum permissible body burden for plutonium-239 is 0.6 μ g. The internal hazards associated with the actinide elements arise from their intense radioactivity (e.g., alpha and beta rays) readily absorbed in the body and the damage done to the body by this radiation originating from internal sites of deposition.

External hazards are those due to the penetrating radiations emitted by the nuclides in question. Generally, only gamma and neutron radiations are considered important in this class, but beta radiation at high intensities must often be considered. The range of alpha radiation in both body tissue and those protective devices normally employed, such as rubber gloves, is sufficiently small to require no consideration at this point. Characteristic X rays of the actinides may be excited in high intensities in the decay. This radiation may make an important contribution to the biological dose when direct manipulation of appreciable quantities of some of the actinides is involved. Fast neutron fluxes are associated with many of the actinide elements as a result of (α ,n) reactions with light elements present as part of the chemical compounds or as impurities. In addition, many of the trans-uranic elements emit fast neutrons by spontaneous fission of some of the

isotopes. The former source of neutrons has only recently received the attention it deserves and a somewhat expanded treatment of this topic is presented. While spontaneous fission also yields fast neutrons, the specific activities are generally low, exceptions being found in certain isotopes of the elements curium, berkelium, californium, and others.

II. THORIUM

Thorium and its compounds have become particularly interesting to the nuclear industry since it became apparent that neutron irradiation of natural thorium, consisting primarily of thorium-232, permits isolation of practical quantities of the fissionable isotope uranium-233. As a result, considerable effort has been expended in the production and final processing of metallic thorium and thorium dioxide components. Many laboratories have become involved in research on the nuclear, physical, and chemical properties of thorium.

The hazards encountered in the handling of thorium can be attributed to 2 sources. First, the decay products of thorium, representing the classical "4n" series, are highly radioactive and constitute both an internal and an external hazard. Secondly, although natural thorium is composed primarily of thorium-232, neutron irradiation of thorium produces additional isotopes as well as changing the ratio of thorium-232 to thorium-228, as normally observed in aged, equilibrium material. A brief review of the decay scheme of thorium-232 as well as the origin and decay data for other isotopes will serve as background for a discussion of the hazards encountered in handling thorium.

Aged natural thorium is composed primarily of thorium-232 in equilibrium with thorium-228. The quantity of thorium-228 in equilibrium with one gram of thorium-232 is $1.35 \times 10^{-4} \mu\text{g}$ or $4.10 \times 10^3 \text{ d/sec}$. Because of the chemical processes during the refining and production of metallic thorium, the radioactive equilibrium is disturbed by removal of the non-thorium daughters. Thus thorium at equilibrium emits 6 alphas, 4 betas and 2.9 gammas per thorium-232 disintegration,^(2,3) while the alpha and gamma activity of freshly separated thorium is slight and no beta radiation is observed. The ratio of alpha radiation from thorium-232 to thorium-228 is approximately unity in freshly separated material. The thorium-228 side chain reaches equilibrium quickly, however, and the alpha and beta activities rise to their first maximum about 36 days after separation. On further aging, the activity decreases, since thorium-228 is removed faster by decay than it is formed from actinium-228. The second maximum in the activity is established at approximately 60 yr after separation. Since these equilibria are disturbed when the daughters are removed by volatilization or chemical processes, it is difficult to estimate the concentration of the daughter products unless detailed information on the history of the material is available. The radiations from thorium are, therefore, closely related to the history of the material and an evaluation of the hazard is most readily obtained by direct measurement.

Natural thorium contains only 2 isotopes; Table 1 presents decay constants⁽⁴⁾ for these and other isotopes of thorium of interest. The gamma radiation in freshly separated thorium is primarily due to the

84-kev radiation from thorium-228. Thorium-230 is a member of the uranium-238 decay chain and is found as a contaminant in thorium separated from uranium ore. Thorium-234 is observed in neutron-irradiated thorium and is formed from thorium-232 by 2 successive neutron-capture reactions.

Table 1

DECAY CONSTANTS OF SELECTED ISOTOPES OF THORIUM⁽⁴⁾

Isotope	Half-life	Specific Activity (d/sec-g)	E_{α} (Mev)	E_{β} (Mev)	E_{γ} (Mev) ^a
Th ²²⁸	1.910 yr	3.04×10^{13}	5.421, 71% 5.338, 28%	-	0.084 (1.6%) 0.132 (1.16%) 0.167 (0.13%) 0.214 (0.30%)
Th ²³⁰	8.0×10^4 yr	7.19×10^8	4.682, 76% 4.615, 24%	-	0.068 (0.59%) 0.142 (0.07%) 0.184 (0.014%) 0.253 (0.016%)
Th ²³¹	25.6 hr	1.96×10^{16}	-	0.299, 39% 0.218, 33% 0.134, 20% 0.090, 8%	0.026 (13%) 0.084 (11%) 0.099 (2%)
Th ²³²	1.39×10^{10} yr	4.10×10^3	4.007, 76% 3.947, 24%	-	-
Th ²³³	22.12 min	1.35×10^{18}	-	1.245, 87%	0.029 (2.1%) 0.087 (2.7%) 0.171 (0.7%) 0.453 (1%)
Th ²³⁴	24.10 d	8.57×10^{14}	-	0.191, 61% 0.103, 35%	0.029 - 0.063 - 0.091 -

^aValues in parentheses are absolute intensities (unconverted photons per 100 disintegrations).

In order to obtain an overall view of the possible changes in the isotopic composition of thorium on irradiation by neutrons, Figure 1 shows the relationships between thorium-232 and the products of neutron irradiation.

The $n,2n$ cross section for thorium-232, which ultimately leads to a rise in the thorium-228 content of irradiated thorium, has been measured by Hasse,⁽⁵⁾ who finds that the ratio of the cross sections for the n,γ and the $n,2n$ processes is 11.3 in an unmoderated fission neutron flux. The main contributors to the hazards associated with irradiated thorium are, therefore, the thorium-234-protactinium-234 chain and the increase in concentration of thorium-228.*

Figure 1

PRODUCTION OF ISOTOPES BY NEUTRON IRRADIATION OF THORIUM-232

The chemical toxicity of thorium is low relative to its radiological toxicity. The insoluble compounds of thorium, such as the dioxide, are chemically remarkably inert to body processes. Ingested thorium is almost completely eliminated with a retention of about $10^{-3}\%$.⁽⁶⁾ The chemical toxicity of thorium dioxide has recently been reported.⁽⁷⁾ Additional references on the chemical toxicity of thorium and its compounds are available.^(2,8)

An appreciation of the magnitude of the radiological toxicity of thorium and its daughters may be obtained by examination of Table 2, which presents a summary of the maximum permissible limits for the several members of the thorium decay chain.⁽⁹⁾ Inspection of the data reveals that the internal hazard from natural thorium is roughly the same as that for plutonium (see footnote c, Table 2, however) and the primary hazard is one of inhalation. The NCRP** has agreed that, until additional data become available, exposure to natural thorium should be kept as low as possible. Hodge⁽¹⁰⁾ has emphasized that no good method of estimating thorium body burdens is available, and therefore the control of atmospheric concentrations of thorium is imperative.

*Protactinium-233, present in high concentration (see Table 5), contributes a major portion of the radiation from freshly discharged and untreated thorium. This element is presumed to be removed in the purification and decontamination treatment.

**National Committee on Radiation Protection.

Table 2

MAXIMUM PERMISSIBLE LIMITS FOR MEMBERS OF THE THORIUM DECAY CHAIN⁽⁹⁾

Nuclide	Critical Organ	MPBB ^a (μ c)	MPC for 40-hr Week ^b	
			Water (μ c/cc)	Air (μ c/cc)
Natural Th ^c	Bone (if soluble)	0.01	3×10^{-5}	(2×10^{-12})
	Lung (if insoluble)	-	-	(4×10^{-2})
	GI	-	3×10^{-4}	5×10^{-8}
Th ^{232c}	Bone (if soluble)	0.04	5×10^{-5}	(2×10^{-12})
	Lung (if insoluble)	-	-	10^{-11}
	GI	-	10^{-3}	2×10^{-7}
Ra ²²⁸	Bone (if soluble)	0.06	8×10^{-7}	7×10^{-11}
	Lung (if insoluble)	-	-	4×10^{-11}
	GI	-	7×10^{-4}	10^{-7}
Ac ²²⁸	GI (if soluble)	-	3×10^{-3}	6×10^{-7}
	Bone	0.04	2	9×10^{-8}
	Liver	0.05	2	8×10^{-8}
	Lung (if insoluble)	-	-	2×10^{-8}
	GI	-	3×10^{-3}	4×10^{-7}
Th ²²⁸	Bone (if soluble)	0.02	2×10^{-4}	9×10^{-12}
	Lung (if insoluble)	-	-	6×10^{-12}
	GI	-	4×10^{-4}	7×10^{-8}
Ra ²²⁴	Bone (if soluble)	0.06	7×10^{-5}	5×10^{-9}
	Lung (if insoluble)	-	-	7×10^{-10}
	GI	-	2×10^{-4}	3×10^{-8}
Rn ^{220d}	Lung	-	-	3×10^{-7}
Pb ²¹⁶	-	-	-	-
Pb ²¹²	Kidney (if soluble)	0.02	6×10^{-4}	2×10^{-8}
	GI	-	6×10^{-4}	10^{-7}
	Lung (if insoluble)	-	-	2×10^{-8}
	GI	-	5×10^{-4}	9×10^{-8}
Bi ²¹²	GI (if soluble)	-	0.01	2×10^{-6}
	Kidney	0.01	0.02	10^{-7}
	Lung (if insoluble)	-	-	2×10^{-7}
	GI	-	0.01	2×10^{-6}
Tl ²⁰⁸	-	-	-	-
Pb ²⁰⁸	-	-	-	-

^aMaximum permissible body burden in total body in microcuries.^bMaximum permissible concentration for a 40-hour week in microcuries per cubic centimeter.^cProvisional values. Although calculations and animal experiments suggest that natural thorium is perhaps as hazardous as plutonium, industrial experience to date has suggested that the hazard of natural thorium is not much greater than that of natural uranium.^dThe daughter isotopes of radon-220 are assumed present to the extent they occur in unfiltered air. For all other isotopes, the daughter elements are not considered as part of the intake and, if present, must be considered on the basis of the rules for mixtures.

The external hazard from natural thorium stems from the gamma-emitting decay products. Roesch⁽¹¹⁾ has calculated the surface dose from massive thorium at equilibrium; his data are shown in Table 3. Roesch also presents data⁽¹¹⁾ which permit calculation of the surface dose rate from thorium at any time after separation. Experimental verification of these calculations showed satisfactory agreement between observed and calculated values. An example of the change of radiation level with time is given by Cuthbert,⁽³⁾ who states that the beta dose rate from one-day-old material is estimated to be less than 10% of the equilibrium dose rate. After 4 days, the radiation is 4 times that of the 1-day-old material. Fifty percent of equilibrium is reached in 10 days.

The formation of radon-220 by the decay of thorium is noteworthy in that relatively large quantities of this isotope are produced. Approximately 0.1 μ c of radon-220 is in equilibrium with one gram of aged, natural thorium. The release of radon is governed by a number of factors, primarily related to the ease with which the gas can diffuse out of and is desorbed from particulates. Chemical and certain physical processes which lead to diminution of aggregate size increase the release of radon into the surroundings. The buildup of radon in confined areas can easily lead to high radiation levels as well as to exceeding of the MPC in air for radon-220 and its daughters. A radon monitor and its application are described by Pradel and Billard,⁽¹²⁾ and considerable information is available on the monitoring of atmospheric radon daughter products.⁽¹³⁾

Table 3

SURFACE DOSE FROM MASSIVE THORIUM AT EQUILIBRIUM⁽¹¹⁾

Nuclide	Radiation	Dose Rate (mrad/hr)	$E\gamma$ (Mev)
Ra ²²⁸ (MsTh ₁)	Beta	1	
Ac ²²⁸ (MsTh ₂)	Beta	36	
Pb ²¹² (ThB)	Beta	10	
	Gamma	10	0.24, 90%
Bi ²¹² (ThC)	Beta	42	
Tl ²⁰⁸ (ThC")	Beta	18	
			2.62, 40%
	Gamma	75	0.58, 40%
			0.51, 20%
	Total	192	
	Total Beta	107	
	Total Gamma	85	

As has been mentioned, the external hazard from irradiated and subsequently purified thorium-232 is primarily due to the thorium-234-protactinium-234 chain and the increase in thorium-228 content of irradiated material from the decay of uranium-232. Arnold⁽¹⁴⁾ indicates that if the thorium-234-protactinium-234 chain is permitted to decay, the thorium-228 chain then becomes the important source of radiation hazard. Thorium-228 is produced by the decay of parasitically produced uranium-232, with decay occurring both during irradiation and during the cooling period following discharge from the reactor. Consequently, the buildup of thorium-228 is related to the total exposure, the number of recycles of thorium, and the cooling times utilized in each cycle. Arnold⁽¹⁴⁾ presents data which will permit estimation of the thorium-228 content of irradiated thorium. An example taken from these data will illustrate the magnitude of the problem. Irradiation of thorium-232 in a flux of 10^{14} n/cm²-sec to a total nvt of 2×10^{21} yields a product containing approximately 6×10^{-3} ppm thorium-228. A billet of such thorium, 10 cm in diameter and 60 cm long, will show a surface dose rate of approximately 8 r/hr and a dose rate of 60 mr/hr at a distance of 1 m. Shielding requirements to reduce the 1-m dose rate to 2 mr/hr are 8 cm of lead or 45 cm of concrete. Cooling for approximately 11 yr will reduce the thorium-228 activity to that of natural thorium. Although these data are rather specific, the implications toward direct processing as well as handling of laboratory samples of irradiated and purified thorium are obvious. The reader is referred to the paper by Arnold⁽¹⁴⁾ for more detailed information.

The formation of fast neutrons by the (α ,n) reaction with light elements is ordinarily not a severe problem when handling thorium, because of its relatively low specific alpha activity. Further, the spontaneous fission half-life of thorium-232 is extremely long ($>10^{20}$ yr), and thus this source of neutrons is unimportant.

In addition to the references cited, Voigt⁽¹⁵⁾ has presented a good summary of the health hazards and radiations of natural thorium, with emphasis on metallurgical manipulations. Ullmann⁽¹⁶⁾ has discussed the effects of buildup of higher isotopes in irradiated thorium.

In summary, the hazards associated with the handling of natural thorium are primarily internal, i.e., inhalation, and are due to the daughters of thorium-232. Natural thorium which has been irradiated in a neutron flux and subsequently purified to remove all protactinium, uranium, and fission products is enriched in thorium-228. This enrichment intensifies the internal hazards as well as produces an appreciable external radiation hazard. No major contribution to the overall hazard is expected from fast neutron emission or the chemical toxicity of thorium.

III. URANIUM

The biological hazards connected with the handling of uranium may be divided into several groups. Uranium-233 presents certain unique problems particularly related to gamma emission because of the nature of the formation of this isotope and the attending contamination from other uranium isotopes. Both natural uranium and uranium-235 may be classed together as a second group, representing a lower gamma hazard when compared with uranium-232-uranium-233. The high specific alpha activity of uranium-232 is more likely to lead to appreciable fast fluxes of neutrons by the (α, n) reaction with light elements. Finally, mention is made of fast neutrons originating from spontaneous fission of several of the uranium isotopes. The decay constants of some pertinent isotopes of uranium are summarized in Table 4 for reference purposes.

Table 4

CONSTANTS OF RADIOACTIVE DECAY FOR SELECTED URANIUM ISOTOPES⁽⁴⁾

Isotope	Half-life	Specific Activity (d/sec-g)	$E_{\alpha, \beta}$ (Mev)	E_{γ} (Mev) ^a
U ²³²	73.6 yr	7.75×10^{11}	5.318, 68%	0.0579 (0.21%)
			5.261, 32%	0.131 (0.75%)
			5.132, 0.32%	0.268 (0.004%)
				0.326 (0.004%)
U ²³³	1.62×10^5 yr	3.50×10^8	4.816, 83.7%	0.029 -
			4.773, 14.1%	0.042 (0.05%)
			4.719, 1.9%	0.056 (0.01%)
			others	0.310 (0.1%)
				others
U ²³⁴	2.48×10^5 yr	2.28×10^8	4.768, 72%	0.053 -
			4.717, 28%	0.118 -
U ²³⁵	7.13×10^8 yr	7.89×10^4	4.559, 6.7%	0.095 (9%)
			4.520, 2.7%	0.110 (5%)
			4.466, 0.9%	0.144 (12%)
			4.369, 29%	0.165 (>4%)
			4.349, 43%	0.185 (55%)
			4.327, 12%	0.205 (4%)
			4.117, 5.8%	
U ²³⁶	2.391×10^7 yr	2.34×10^6	4.499, 73%	0.163 (0.5%)
			4.45, 27%	
			4.337, 0.5%	
U ²³⁷	6.75 d	3.02×10^{15}	0.248, 74% α^-	0.060 (38%)
			0.084, 26% β^-	0.114 -
				0.163 (3.6%)
				0.208 (24%)
				0.266 (0.9%)
				0.332 (2%)
U ²³⁸	4.51×10^9 yr	1.23×10^4	4.195, 77%	0.048 -
			4.147, 23%	

^aValues in parentheses are absolute intensities (number of unconverted photons per 100 disintegrations).

Uranium produced by the neutron irradiation of thorium-232 is normally composed of several isotopes. The isotopic composition is not readily predictable unless several important reactor parameters are specified and, in fact, uncertainties in the fundamental data such as neutron cross sections limit the utility of exact calculations. An example of the calculations which have been carried out is that of Schuman and Tromp,⁽¹⁷⁾ who have calculated the composition of reactor-irradiated thorium-232 as well as uranium-233 for neutron energy distributions found in the MTR.* Table 5 shows an example of the variation of the isotopic composition of pile-irradiated thorium, and Table 6 presents similar data for the irradiation of uranium-233. The data in Tables 5 and 6 are illustrative of the magnitude of the concentrations of the several isotopes of uranium and are used in examples of radiation calculations. Arnold *et al.*^(18,19) have calculated the isotopic composition of uranium produced by thermal neutron irradiation of thorium. The data are presented graphically for several values of the neutron flux and may be used to estimate the isotopic composition of uranium obtained from thermal neutron irradiation of thorium in a homogeneous⁽¹⁸⁾ and a heterogeneous⁽¹⁹⁾ reactor.

Table 5

COMPOSITION OF PILE-IRRADIATED THORIUM⁽¹⁷⁾

Thermal Flux = 2×10^{14} n/cm²-sec
 Resonance Flux = $\frac{1}{12}$ Thermal Flux
 Fast Flux = Resonance Flux

nvt (n/cm ²)	time (days)	Atoms per 10 ⁶ Initial Th ²³² Atoms					
		Th ²³²	Pa ²³³	U ²³²	U ²³³	U ²³⁴	U ²³⁵
4×10^{19}	2.32	999,420	562	0.002	16.5	1.04	0.003
2×10^{20}	11.6	997,102	2,489	0.048	366	25.2	0.262
8×10^{20}	46.3	988,457	6,591	0.684	3,840	357	13.7
2×10^{21}	116	971,392	8,724	3.40	11,395	1,718	144
8×10^{21}	463	890,386	8,381	19.5	18,031	8,529	1,597
2×10^{22}	1160	748,077	7,041	26.1	15,271	11,169	2,442

*Materials Testing Reactor.

Table 6

COMPOSITION OF IRRADIATED $U^{233(17)}$

$$\text{Resonance Flux} = \frac{1}{12} \text{ Thermal Flux}$$

nvt^a (n/cm^2)	Atoms per 10^6 Initial U^{233} Atoms				
	U^{233}	U^{234}	U^{235}	U^{236}	$U^{237} + Np^{237}$
4×10^{19}	972,777	2,871	9.16	0.015	0
2×10^{20}	871,099	13,417	210	1.61	0
8×10^{20}	575,797	41,879	2,465	81.3	0.534
2×10^{21}	251,579	65,365	8,429	809	13.7
8×10^{21}	4,006	37,744	10,002	7,939	562
2×10^{22}	1.02	5,614	1,570	10,326	1,442

^aThe values depend only on "nvt," except $U^{237} + Np^{237}$ which varies slightly with flux.

By means of the data of Tables 5 and 6 as a guide, a rough evaluation of the source and approximate magnitudes of the hazards may serve to illustrate the problems encountered in handling uranium-233. It should be emphasized that evaluation of the specific problems is best made by measurement of the samples in question. The internal hazard from uranium obtained by irradiation of thorium-232 and subsequent chemical purification arises from the decay of the short-lived uranium-232 through the thorium-228 chain discussed previously as well as some contribution from uranium-233 and its daughters. Although the internal hazard from thorium-228 and its daughters is appreciable, their concentrations are generally rather low, and the major problems are usually external, arising from beta, gamma, and neutron radiation from uranium-232 and its decay products. Devaney⁽²⁰⁾ has calculated the gamma-ray intensity at the surface of nickel-coated uranium spheres of varying radii and containing variable amounts of uranium-232. Correction factors for the growth of the uranium-232 daughters have been included.

Devaney's data⁽²⁰⁾ are presented in the form

$$D = AKC \quad ,$$

where D is the dose rate in units of r/hr at the surface of a sphere of uranium of radius r, containing C parts uranium-232 per million parts uranium. Values for A are given below.

<u>Radius of Sphere r, cm</u>	<u>A</u>
2	14.0
3	14.8 ₇
4	15.0 ₄
5	15.2 ₃

The value of K is obtained from the equation

$$K = e^{-0.0099t} - e^{-0.3647t} \quad (t \text{ in years})$$

Arnold⁽²¹⁾ has issued a useful tabulation of the buildup and decay of members of the uranium-232 decay chain, which permits estimation of the activity of each member of the chain as a function of time.

Arnold⁽¹⁴⁾ has examined the problems of handling uranium-233 contaminated by uranium-232 from the standpoint of processing and fabricating recycled power reactor fuel. Arnold presents data which relate the uranium-232 content to irradiation history expressed in terms of nvt and the gamma dose rates per ppm uranium-232 at a distance of 1 m from a 1-kg sphere of uranium-233, tabulated for varying aging times. Although little indications are given as to the bases for the calculations, agreement between the data of Arnold and those of Devaney⁽²⁰⁾ is adequate. Table 7 presents a selection of data calculated by Arnold,⁽¹⁴⁾ showing the rapid rise in dose rate with time after separation. Lawrence⁽²²⁾ has calculated data similar to those of Arnold⁽¹⁴⁾ and has included an extensive discussion of problems of air-borne activity and maximum permissible body burdens. Data for small samples of uranium-233 contaminated with uranium-232 and its daughters are also included. Aside from the calculated intensities mentioned above, certain generalities are worth noting. The surface gamma dose rates, even from small but aged pieces of uranium-233 contaminated with uranium-232, may be considerable. Further, increasing the state of subdivision of the material produces profound changes in the handling hazard of laboratory-size samples by increasing the apparent beta dose rate as well as from the release of radon-220. The beta dose from uranium-233 (and thorium) is primarily due to bismuth-212 (2.25 Mev) and thallium-208 (1.8 Mev), which also contribute highly energetic gamma rays. As was mentioned previously, the release of radon-220 may create an internal hazard as well as an external one and deserves careful consideration and attention. Further appreciation of the precautions utilized in the handling of uranium-233 may be gained by examination of the compendium edited by Walton.⁽²³⁾

Table 7

RADIATION INTENSITIES AT 1 METER FROM A 1-KG SPHERE OF U²³³ CONTAINING U²³²⁽¹⁴⁾

<u>Time after Separation (sec)^a</u>	<u>1 x 10⁴</u>	<u>1 x 10⁵</u>	<u>1 x 10⁶</u>	<u>1 x 10⁷</u>	<u>3 x 10⁷</u>	<u>6 x 10⁷</u>
<u>Dose Rate per ppm U²³² (mr/hr)</u>	3.2 x 10 ⁻⁷	5.7 x 10 ⁻⁴	0.0835	1.38	3.84	6.55

^a1 yr = 3.156 x 10⁷ sec

Highly enriched uranium, containing primarily uranium-235, presents the normal problems attending the $4n + 3$ decay chain. Protactinium-231, actinium-227, and thorium-227 are members of the decay chain having rather low ($<10^{-10} \mu\text{c}/\text{cm}^3$) MPC values in air for a 40-hr/week exposure. No data on the surface dose rates from massive samples of uranium-235 appear to have been published.

Bortner and Richards⁽²⁴⁾ report observations on the surface beta dose from enriched uranium (27.5% U^{235}) and thorium. These authors found satisfactory agreement between the calculated and observed ratio of the air dose from natural and enriched uranium. The observed air dose for natural uranium is given by Bortner,⁽²⁵⁾ who measured the dose rate from a thick disk of metal with an extrapolation chamber. The total air dose, including that from alpha radiation, was observed to be 14.4 esu/cc-hr. When the alpha radiation was filtered by 6.9-mg/cm² polystyrene, the residual dose rate, primarily from beta radiation, was 0.24 rep/hr. Bass *et al.*⁽²⁶⁾ report measurements of the surface beta dose from metallic uranium as well as from uranium dioxide, uranium trioxide, U_3O_8 , uranium tetrafluoride, uranyl nitrate hexahydrate, and sodium diuranate. The data for metallic uranium are in agreement with those of Bortner⁽²⁵⁾ mentioned above.

Neutron irradiation of uranium-235 and subsequent purification of the uranium to remove fission products and other impurities results in the buildup of other isotopes of uranium. Specifically, uranium-236 and uranium-237 will be produced by neutron-capture reactions. The isotopic composition of irradiated uranium will vary depending on the characteristics of the neutron flux, the total exposure, and the magnitude of the flux.⁽¹⁷⁾ Arnold⁽¹⁴⁾ has pointed out that the primary external hazard associated with irradiated and purified uranium stems from the decay of uranium-237, which proceeds by beta emission accompanied by several gamma rays. A complication is introduced by consideration of the extent of the removal of uranium-236 in the isotope-separation scheme used to re-enrich the irradiated uranium-235, since the degree to which uranium-236 is removed influences the level of uranium-237 produced by subsequent neutron irradiation. A discussion of this aspect is also presented by Arnold.⁽¹⁴⁾

The gamma-activity level of uranium-237 is high because of its short (6.75-day) half-life and the profusion of gamma rays which accompany its decay. Arnold⁽¹⁴⁾ has calculated that the surface dose rate from a spherical 1-kg sample of uranium containing 1 c (12.2 μg) of uranium-237 is 5.8 r/hr. Although the internal hazards from the decay products of uranium-235, uranium-236, and uranium-237 are considerable, the external hazards from the gamma rays appear to warrant considerably more immediate attention in this case. In the case of uranium-235 (containing uranium-237) as in most other instances, the specific problems are best evaluated by direct measurements made on the specific sample.

Depleted uranium or uranium containing primarily uranium-238 presents problems similar to those discussed for uranium-238. One gram of aged uranium-238 contains $0.33 \mu\text{c}$ of uranium-234 and its decay products, which include thorium-230, radium-226, and radon-222, all of which have relatively low MPBB and MPC values for air.⁽⁹⁾ The surface gamma dose rate for natural uranium is stated⁽¹⁴⁾ to be 30 mr/hr which, while relatively low, is considerably greater than the 2.5 mr/hr (100 mr/40-hr week) set for the maximum permissible occupational exposure.⁽²⁷⁾ A report by Redfearn and Mott⁽²⁸⁾ describes the measurements made with ingots of alloyed uranium. The observed increase in surface beta activity is believed due to segregation at the surface of the thorium isotopes from the decay of uranium. The increase in surface beta-gamma counts amounted to a factor of 12 in several instances. As in the case of uranium-235, irradiation of uranium-238 by neutrons leads to the formation of uranium-237 by the $\text{U}^{238} (n, 2n) \text{U}^{237}$ reaction. The hazards connected with the handling of uranium-238 are thus also related to the history of the material and, although the absence of appreciable concentrations of uranium-237 sharply diminishes the problem, evaluation of the actual samples appears to be the most direct approach. The beta dose from normal uranium comes primarily from protactinium-234 (1.50- and 2.32-Mev maximum beta energy). Arnold⁽¹⁴⁾ has tabulated the beta surface dose rates for beta energies encountered in uranium-232, uranium-233, and natural uranium.

Whereas gamma radiation represents a considerable external hazard in many instances, neutron emission from several sources may represent an important contribution to the total external hazard. Fast neutrons are produced by 2 important sources in samples of uranium. Spontaneous fission of several isotopes contributes a small neutron flux from samples of uranium, particularly if the samples contain appreciable quantities of uranium-232. In addition, (α, n) reactions with light elements present as either impurities or as integral parts of the compounds in question also serve as a source of fast neutrons.

Harris⁽²⁹⁾ has issued a valuable review of the production of neutrons from spontaneous fission, (α, n) reactions, and other sources. Table 8 lists the decay constants for spontaneous fission of several uranium isotopes, taken primarily from the compendium of Strominger *et al.*⁽³⁰⁾ Natural uranium shows a neutron yield from spontaneous fission of 1.6×10^{-2} n/sec-g,⁽²⁹⁾ whereas enriched uranium, containing 93.2% U^{235} , 1% U^{234} , 0.3% U^{236} , and 5.5% U^{238} , exhibits a neutron emission rate from spontaneous fission of 1.5×10^{-3} n/sec-g. These values are relatively small when evaluated in terms of a biological hazard, particularly in the light of the approximate relation $1 \text{ nf/cm}^2\text{-sec} \equiv 0.14 \text{ mrem/hr.}^*$

*Based on 1.0-Mev neutrons, $18 \text{ nf/cm}^2\text{-sec} = 100 \text{ mrem/40-hr week.}^{(33)}$

Table 8

SPONTANEOUS FISSION DECAY CONSTANTS OF
URANIUM ISOTOPES⁽³⁰⁾

<u>Isotope</u>	<u>Half-life (yr)</u>	<u>λN (d/sec-g)</u>	<u>$\bar{\nu}$ (n/dis)</u>
U ²³²	$\sim 8 \times 10^{13}$	0.71	-
U ²³³	$> 2 \times 10^{17}$	$< 2 \times 10^{-4}$	-
U ²³⁴	2×10^{16}	2.85×10^{-3}	$\sim 1.7^a$
U ²³⁵	1.9×10^{17}	2.94×10^{-4}	$\sim 1.7^a$
U ²³⁶	2×10^{16}	2.85×10^{-3}	$\sim 1.5^a$
U ²³⁸	1×10^{16}	5.55×10^{-3}	2.1^b

^aReference 29

^bKuzimov, B. D., et al., J. Expt. and Theor. Phys. (USSR) 37, 2, 8 (1959).

Arnold⁽¹⁴⁾ has tabulated the dose rates expected from spontaneous fission neutrons for one-kilogram spheres of uranium-233 contaminated with uranium-234 and small quantities of uranium-232, as well as the dose rates from uranium-235 and uranium-238. Although the method of calculation is not detailed, in all cases the surface flux of fast neutrons is less than 0.5 n/cm²-sec and the surface dose rates are less than 0.1 mr/hr.

In contrast with the relatively low biological hazards from neutrons arising by spontaneous fission of uranium isotopes, neutrons produced by (α , n) reactions may represent a severe hazard. Harris⁽²⁹⁾ has summarized the available data for certain light elements and has calculated values of the yield integral:

$$\int_0^{E_0} \frac{a}{A_j} \sigma_j \left(-\frac{dx}{dE} \right) dE \quad ,$$

where a is Avogadro's number, A_j is the atomic weight of the j th component, σ_j is the microscopic (α , n) cross section, and $-dx/dE$ is the inverse rate of energy loss of alpha particles in the medium. Values have been calculated for Al, Mg, B, O, Na, C, and Be in media important in reactors. Thick target yields for B, Be, and F have been measured by Segre and Wiegand⁽³¹⁾ as a function of alpha-particle energy, and Runnalls⁽³²⁾ has determined the thick target yield of beryllium as a function of alpha energy. These data permit estimation of the neutron emission rates of compounds of uranium containing alpha-emitting isotopes of high specific activity. Table 9 presents

a summary of the calculated neutron emission rates from selected compounds of uranium. Although the data in Table 9 are calculated and thus subject to the limitations of the assumptions, they clearly illustrate the magnitude of the neutron flux. Arnold⁽¹⁴⁾ has collected data applicable to the U^{232} - U^{233} - Th^{228} system; these permit ready estimation of the neutron emission rates from uranium containing light elements. Arnold has combined estimates of the yield integral and specific activity data so that specific neutron emission rates may be derived from a simple three-constant equation. Values for the three constants are tabulated⁽¹⁴⁾ for all pertinent elements from lithium to nickel.

Table 9

CALCULATED NEUTRON EMISSION RATES FOR
SELECTED COMPOUNDS OF URANIUM

<u>Compound</u>	<u>n/sec-g U^a</u>	<u>ϕ (n/cm²-sec)^b</u>
$U^{232}F_4$	1.86×10^6	1.23×10^7
$U^{233}F_4$	4.2×10^2	2.78×10^3
$U^{235}F_4$	3.8×10^{-2}	0.25
$U^{238}F_4$	5.9×10^{-3}	3.9×10^{-2}
$U^{232}O_2$	1.86×10^4	2×10^5
$U^{233}O_2$	5.9	63
$U^{235}O_2$	1.04×10^{-3}	1.1×10^{-2}
$U^{238}O_2$	1.48×10^{-4}	1.6×10^{-3}

^aThe following integral yields were used: ($\times 10^{+6}$)

U^{232} : F - 10, 0 - 0.20; U^{233} : F - 5, 0 - 0.14

U^{235} : F - 2, 0 - 0.11; U^{238} : F - 2, 0 - 0.10

^bFast neutron flux passing through a sphere of unit cross-sectional area at the surface of a 500-g sphere of the indicated compound, compacted to its crystal density. The flux is related to the radius of the sphere, r , and the specific volume neutron emission rate a (n/cc-sec) by $\phi = \frac{1}{2} ar$. We are indebted to Dr. W. C. Roesch for pointing out this relationship. [See also Whyte, G. N., Principles of Radiation Dosimetry, J. Wiley and Sons, Inc., New York (1959) Ch. 1.]

A brief comment on neutron energies is necessary in order to permit proper evaluation of the hazards from this source. Little if any experimental data are available for the energy distribution of neutrons emitted by (α , n) sources when the alpha-active component is uranium, and, although it is possible to calculate an energy distribution, the need for such calculation is normally absent. If the assumption is made that all neutrons have an energy of 1 Mev (maximum RBE* of 10.5), then the dose obtained per unit flux is 0.14 mrem/hr.⁽³³⁾ Thus, 1 n/cm²-sec = 0.14 mrem/hr, and the maximum permissible flux which will deliver 100 mrem in a 40-hr week is 18 n/cm²-sec. Any error introduced by this assumption will increase the maximum permissible neutron flux. Since shielding materials such as water or other hydrogenous materials are also moderators, they alter the energy spectrum of the neutron flux, and measurements made on neutron sources should include provisions for differentiating between slow and fast neutrons, since the RBE and the values for the maximum permissible neutron flux are dependent on the neutron energy.⁽³³⁾

*RBE = Relative Biological Effectiveness

IV. PLUTONIUM

Plutonium is perhaps the most important of several artificially produced elements, because of the military uses which have involved this element. The handling of plutonium and its compounds represents, therefore, a well-established, albeit restricted, technology, and considerable information on the handling of plutonium is available.

The advent of power reactors fueled with natural or slightly enriched uranium marks the beginning of the production of large commercial quantities of plutonium. It has been estimated* that by the end of the 1960's the United Kingdom will produce from its commercial reactors about 5 tons of plutonium per year which, if not utilized, would represent a waste of about $\$7.5 \times 10^7$ per yr at a plutonium price of \$15 per g. A parallel situation may be envisioned for the United States. Thus, increasing pressure toward utilization of plutonium in reactors and other devices appears inevitable, and, indeed, creation of facilities such as the Argonne Plutonium Fuel Fabrication Facility indicates awareness of this problem. Reactor physics studies are also taking cognizance of this problem and, for example, Okrent⁽³⁴⁾ has considered the effect of the isotopic compositions of the plutonium feed material on the breeding ratio and other parameters. A marked increase in research and development and the attending handling of plutonium may therefore be safely anticipated. It is in this connection that an evaluation of the hazards in the handling of plutonium becomes particularly appropriate.

The internal hazard from plutonium is paramount when relatively small quantities of the material are involved and arises principally because of the high specific alpha activity of most of the isotopes. The external hazard from plutonium is due to the gamma and fast neutron emission of several of the isotopes and is therefore a function of the isotopic composition of the sample. In order to evaluate the hazards from a sample of plutonium, a knowledge of the isotopic composition is required, together with data about the decay characteristics of the several isotopes.

Plutonium is produced in reactors by neutron-capture reactions in uranium. Figure 2 illustrates the important nuclear reactions leading to the formation of plutonium. The dependence of the cross sections for each of the neutron-capture reactions on the neutron energy leads to a strong dependence of the isotopic composition of the plutonium on the reactor characteristics and initial fuel composition. Since it is not possible, therefore, to predict in a simple manner the isotopic composition of a sample of

*Sir John Cockcroft in the dedication address for the Plutonium Fuel Fabrication Facility, Argonne National Laboratory, May 14, 1959.

plutonium from exposure alone, the data of Schumann and Tromp,⁽¹⁷⁾ while quite specific, are again chosen to illustrate the variations which may be found. It needs to be pointed out that these data are not typical of the isotopic composition of plutonium from power reactors, and a direct determination of these ratios is required for a hazards evaluation pertaining to a specific sample of plutonium.

Figure 2

FORMATION OF PLUTONIUM ISOTOPES FROM URANIUM IN NUCLEAR REACTORS

Table 10, taken from the data of Schumann and Tromp,⁽¹⁷⁾ illustrates the variation of the isotopic composition of plutonium obtained by irradiation of uranium-238.

Table 10

COMPOSITION OF PLUTONIUM OBTAINED BY IRRADIATION OF URANIUM-238⁽¹⁷⁾

Resonance Flux per In E Interval = 1/30 Thermal Flux
Fast Flux = 1/3 Thermal Flux

nvt (n/cm ²)	Total Pu ^a (Atoms/10 ⁶ Initial U238)	Fraction of Total Plutonium				
		Pu238	Pu239	Pu240	Pu241	Pu242
$\phi_{th} = 5 \times 10^{13}$ n/cm ² -sec						
5 x 10 ¹⁹	596.7	0.17 x 10 ⁻⁶	0.9939	6.08 x 10 ⁻³	5.19 x 10 ⁻⁵	0
2 x 10 ²⁰	2,250	3.2 x 10 ⁻⁶	0.9665	0.0323	1.15 x 10 ⁻³	2.18 x 10 ⁻⁵
2 x 10 ²¹	12,931	136 x 10 ⁻⁶	0.6917	0.234	0.058	0.016
1 x 10 ²²	21,859	4340 x 10 ⁻⁶	0.406	0.262	0.098	0.230
$\phi_{th} = 2 \times 10^{14}$						
2 x 10 ²²	24,571	11.8 x 10 ⁻⁶	0.336	0.209	0.080	0.374

^aAssuming all Np239 has decayed to Pu239 but no loss of plutonium by decay

Similar data are available⁽¹⁷⁾ for uranium-235.* Although the example of Table 10 is only illustrative of the variations which are produced on extended irradiation, considerable information is available which pertains more directly to power reactor fuels. In a subsequent portion of this discussion, reference to more practical data will be made to illustrate the radiation hazards from irradiated power reactor fuels.

The decay characteristics of the important plutonium isotopes are shown in Table 11.

Table 11

DECAY CONSTANTS FOR SELECTED ISOTOPES OF PLUTONIUM⁽³⁰⁾

Isotope	Half-life (yr)	Specific Activity (d/sec-g)	$E_{\alpha, \beta}$ (Mev)	E_{γ} (Mev) ^a
<u>Alpha Decay</u>				
Pu ²³⁸	86.4	6.43×10^{11}	5.495, 72%	0.013 (0.038%)
			5.452, 28%	0.099 (0.009%)
				0.150 (0.001%)
Pu ²³⁹	2.436×10^4	2.27×10^9	5.147, 72.5%	0.038 (0.002%)
			5.134, 16.8%	0.052 (0.007%)
			5.096, 10.7%	0.120 (0.002%)
Pu ²⁴⁰	6.58×10^3	8.38×10^9	5.162, 76%	0.045 -
			5.118, 24%	
Pu ²⁴¹	13.3	4.12×10^{12}	0.0208 β	
	5.72×10^5 (36)	9.6×10^7	4.9 α	0.145 (7%)
Pu ²⁴²	3.79×10^5	1.44×10^8	4.898, 76%	
			4.858, 24%	
<u>Spontaneous Fission</u>			<u>$\bar{\nu}(n/d)$(35)</u>	
Pu ²³⁸	3.8×10^{10}	1.45×10^3	2.33	
Pu ²³⁹	5.5×10^{15}	1.0×10^{-2}	(2.3) ^b	
Pu ²⁴⁰	1.22×10^{11}	4.52×10^2	2.25	
Pu ²⁴²	7.1×10^{10}	7.8×10^2	2.18	

^aValues in parentheses are absolute intensities, i.e., unconverted photons per 100 disintegrations.

^bEstimated.

*Okrent (Ref. 34) used pure plutonium-239; plutonium composed of 74.7% Pu²³⁹, 10.2% Pu²⁴⁰, 12.4% Pu²⁴¹, and 2.7% Pu²⁴²; and plutonium composed of 40% Pu²³⁹, 10% Pu²⁴⁰, 25% Pu²⁴¹, and 21% Pu²⁴² for his calculations. Arnold⁽¹⁴⁾ considered plutonium having the composition 36.15% Pu²³⁹, 14.71% Pu²⁴⁰, 8.82% Pu²⁴¹, and 40.31% Pu²⁴² as "equilibrium" plutonium.

The average number of neutrons per spontaneous fission is taken from the data of Hicks *et al.*⁽³⁵⁾ Data for plutonium-241 are from Brown *et al.*⁽³⁶⁾ Inspection of the data of Table 11 shows that, as the concentration of the higher isotopes in plutonium rises, a rise in specific alpha activity as well as an increase in the neutron emission rate from spontaneous fission can be anticipated. Whereas the former becomes important only when considering (α, n) reactions, the latter is independent of the impurity level or nature of the compound.

The magnitude of the internal hazard, i.e., the toxicity of plutonium, may be estimated from the data in Table 12 which lists the maximum permissible limits for several plutonium isotopes.⁽⁹⁾ The internal hazard from plutonium as evidenced by the data in Table 12 is normally considerably greater than the external one, and much of the work with plutonium and its compounds is confined to special enclosures and glove boxes. The interested reader is referred to several references for additional information^(23,37,38) on this topic.

Table 12

MAXIMUM PERMISSIBLE BODY BURDENS AND MAXIMUM PERMISSIBLE
CONCENTRATIONS OF PLUTONIUM IN AIR AND WATER⁽⁹⁾

Isotope	Form	Critical Organ	MPBB ^a		MPC ^b per 40-hr week	
			(μ c)	(μ g)	Water (μ c/cc)	Air (μ c/cc)
Pu ²³⁸	Sol	Bone	0.04	2.4×10^{-3}	10^{-4}	2×10^{-12}
Pu ²³⁹	Sol	Bone	0.04	0.65	10^{-4}	2×10^{-12}
	Insol	Lungs ^c	0.008	0.13	-	2×10^{-12}
Pu ²⁴⁰	Sol	Bone	0.04	0.18	10^{-4}	2×10^{-12}
Pu ²⁴¹	Sol	Bone	0.9	8.2×10^{-3}	7×10^{-3}	9×10^{-11}
Pu ²⁴²	Sol	Bone	0.05	12.8	10^{-4}	2×10^{-12}

^aMaximum permissible total body burden in μ c or μ g.

^bMaximum permissible concentration in μ c/cm³.

^cThe low value for insoluble plutonium-239 is quoted in an earlier edition of reference 9 (Handbook 52, 1953) and is still used even though it is not included in reference 9.

Even though the internal hazard from plutonium may be minimized by use of special enclosures, the external hazard may not be negligible, particularly when larger quantities of plutonium from power reactor fuels are considered. The external hazards from plutonium have been evaluated by Roesch,⁽³⁹⁾ Birchall,⁽⁴⁰⁾ and Arnold.⁽¹⁴⁾ Roesch and Birchall have calculated the surface dose contributed by each of the isotopes of plutonium likely to be encountered in power reactor fuels. Although the method of calculation used by each of these authors is different, the results appear to be comparable, particularly in the light of the difficulty of the problem. A comparison of the data is given in Table 13. The data in Table 13 are for spherical samples of massive plutonium, and Birchall lists conversion factors applicable to other shapes. Arnold⁽¹⁴⁾ has calculated the radiation intensities from cylindrical shapes of metallic plutonium-239 and

plutonium-240 for various cylinder radii, heights, and distances from the cylinder. Arnold has further divided the dose rate into that from soft radiation (L X rays) and that from hard radiation (K X and gamma rays). This division allows estimation of dose rates when handling plutonium under normal glove box conditions.

Table 13

SURFACE DOSE RATES FROM MASSIVE PLUTONIUM

Isotope	Radiation and Energy	Surface Dose Rate (rad/hr)	
		Roesch(39)	Birchall(40)
Pu ²³⁸	X ray (~20 kev)	980	736
	Gamma ~45 kev	2	6.95
	150 kev	-	1.87
	100 kev	-	13.3
		<hr/> 982	<hr/> 758
Pu ²³⁹	X ray (~20 kev)	0.61	0.751
	Gamma (several)	0.056	-
	38 kev	-	0.001
	52 kev	-	0.006
	120-207 kev	-	0.013
	380 kev	-	0.302
		<hr/> 0.666	<hr/> 1.073
Pu ²⁴⁰	X ray (~20 kev)	14	7.2
	Gamma 44 kev	0.008	0.02
	1 Mev (spont. fiss.)	0.37	-
		<hr/> 14.38	<hr/> 7.22
Pu ^{241a}	Gamma 100 kev	-	10.04
	145 kev	-	2.37
			<hr/> 12.37
Pu ²⁴²	X ray (~17 kev)	-	0.06
Am ^{241b}	X ray 17 kev	742.5 ^c	-
	26 kev	22.7	33
	43 kev	6.5	-
	Gamma 60 kev	713	2680
	99 kev	16	-
		<hr/> 1483 ^c	<hr/> 2713
U ^{237d}	Beta 250 kev	-	3.0 x 10 ⁸
	Gamma 60 kev	-	0.61 x 10 ⁸
	210 kev	-	5.91 x 10 ⁸
	330 kev	3.92 x 10 ^{8c}	1.77 x 10 ⁸
		<hr/> 3.92 x 10 ⁸	<hr/> 11.3 x 10 ⁸

^aArnold(14) estimates 20-30 r/hr from a 1-kg sphere of pure Pu²⁴¹.

^bThe growth of Am²⁴¹ from Pu²⁴¹ can be approximated by the equation(39) Am²⁴¹ (curies/g of initially pure Pu²⁴¹) = $4.28 \times 10^{-4}t$ for t (days) < 14 years.

^cRoesch assumed a source density of 1 c/g. The data have been converted where appropriate to obtain comparable figures.

^dThe growth of U²³⁷ from Pu²⁴¹ can be approximated by the equation(39) U²³⁷ (curies/g of initially pure Pu²⁴¹) = $4.7 \times 10^{-3} [1 - \exp(-0.102t)]$ for t (days) < 14 years.

Data on the experimentally determined surface dose rate from massive plutonium are available from the work of Schiavone *et al.*⁽⁴¹⁾ and data quoted by Dummer.⁽⁴²⁾ Although both of these references fail to specify the isotopic composition of the plutonium, Schiavone finds a total surface dose rate of 10.4 rep/hr from beta and gamma components. The dose rate through 9 to 11 mils (22-27 mg/cm²) of latex gloves together with 30 to 35 mils (105-122 mg/cm²) of neoprene (dry box) gloves was found to be 490 mrep/hr. Dummer quotes a value of 360 mrad/hr to the bare hand from nickel-coated plutonium and 440 mrad/hr from uncoated plutonium through dry box gloves. The gamma spectrum of plutonium tetrafluoride has been reported⁽⁴³⁾ and shows peaks at 17, 30, 60, 100, 130, 160, 200, 380, 500, 850, 1200, 1700, and 1950 kev. These data are in qualitative agreement with those reported by Moulthrop,⁽⁴⁴⁾ who also tabulated data about the gamma spectrum of metallic plutonium, plutonium oxalate, plutonium oxide, and plutonium nitrate.

Neutrons from plutonium and its compounds are produced by spontaneous fission and by (α ,n) reactions with light elements. The spontaneous fission half-lives and decay constants are shown in Table 11. These data permit estimation of the contribution of spontaneous fission to the total neutron flux from samples of plutonium. These estimates are included in Table 14.

Data on the yield integral for neutron emission from light elements due to the (α ,n) reaction are generally not available. The data of Segre and Wiegand⁽³¹⁾ and Roberts⁽⁴⁵⁾ have been used to estimate the yields shown in Table 14. A survey by Hess⁽⁴⁶⁾ gives data on the energy spectrum of neutrons from several sources, including Po-F and Pu-Be. Although no data on Pu-F and Pu-O are available, it can readily be shown that the neutron energies are in the "fast" region, and estimates of the mean energy vary from 0.75 Mev to 1.5 Mev. In order to appreciate the magnitude of the neutron flux expected from samples of plutonium fluorides and oxides, Table 14 summarizes calculations for these compounds with the neutrons from spontaneous fission. Arnold⁽¹⁴⁾ has tabulated constants for elements from lithium to iron suitable for calculating the neutron emission rates from a simple two-constant equation. The effect of plutonium-240 and plutonium-238 is included in the equation given by Arnold.⁽¹⁴⁾

Experimental data on the neutron flux from samples of plutonium fluorides (or plutonium metal) are not generally available. Helgeson⁽⁴⁷⁾ has indicated that a 450-g sample of PuF₄ contained in a $\frac{1}{4}$ -in.-wall Plexiglass container has a surface dose rate of 3.0 rem/hr fast neutrons and 1.5 rem/hr gamma and X ray. The average energy of the neutrons is stated to be 0.75 Mev. Although no primary data are given in this report, a surface dose rate of 3.0 rem/hr of 0.75-Mev neutrons is approximately equivalent to a specific neutron emission rate of 5300 n/sec-g Pu. This value compares favorably with those calculated for Pu²³⁹F₄ and Pu²⁴⁰F₄, shown in Table 14.

Table 14

FAST NEUTRON EMISSION FROM METALLIC PLUTONIUM
AND SELECTED COMPOUNDS

Substance	n/sec-g Pu		ϕ from 500-g Sphere ^c (n/cm ² -sec)	
	(α, n) ^a	Spontaneous Fission ^b	(S.F.) ^d	(α, n)
Pu ²³⁸	-	3.4×10^3	6.1×10^4	-
Pu ²³⁸ F ₄	2.1×10^6	-	2.3×10^4	1.4×10^7
Pu ²³⁸ O ₂	1.4×10^4	-	3.3×10^4	1.3×10^5
Pu ²³⁹	-	0.02	0.4	-
Pu ²³⁹ F ₄	4.3×10^3	-	0.13	2.9×10^4
Pu ²³⁹ O ₂	45	-	0.20	4.4×10^2
Pu ²⁴⁰	-	1×10^3	1.8×10^4	-
Pu ²⁴⁰ F ₄	1.6×10^4	-	6.9×10^3	1.1×10^5
Pu ²⁴⁰ O ₂	170	-	1×10^4	1.6×10^3
Pu ²⁴²	-	1.7×10^3	3.1×10^4	-
Pu ²⁴² F ₄	1.7×10^2	-	1.1×10^4	1.2×10^3
Pu ²⁴² O ₂	2.7	-	1.7×10^4	27

^aThe following yield integral values were used: ($\times 10^6$)

Pu²³⁸: F - 14; 0 - 0.18; Pu²³⁹: F - 8; 0 - 0.17;

Pu²⁴⁰: F - 8; 0 - 0.17; Pu²⁴¹: F - 5; 0 - 0.16.

^bSee Table 11 for constants.

^cFast neutron flux at the surface of a 500-g sphere of the indicated compound at its crystal density, passing through a sphere of unit cross-sectional area (See footnote b, Table 9).

^dSpontaneous Fission.

Plutonium containing appreciable quantities of plutonium-241 as well as other isotopes is obtained from highly irradiated uranium such as that discharged from power reactors. While plutonium-241 decays primarily by beta emission to americium-241, a small fraction of the decays are by alpha-particle emission with the formation of uranium-237. Birchall⁽⁴⁰⁾ has shown that the activity of uranium-237 in plutonium from power reactors can be expected to reach equilibrium in 6 to 8 weeks, and the contribution of uranium-237 to the total surface dose may be estimated from the data

in Table 11. Americium-241, on the other hand, continues to build up for considerable periods after separation and, in order to assess the contribution from this isotope, the time after separation must be known. As has been mentioned, the exact isotopic composition of plutonium discharged from power reactors is dependent on a number of interrelated factors. It is instructive, however, to note the changes which occur in the surface dose rates and the neutron emission rates from discharged plutonium as the in-pile exposure increases. The data of Birchall⁽⁴⁰⁾ are used as an example, but Roesch⁽³⁹⁾ and Arnold⁽¹⁴⁾ present similar data. A summary of the data is shown in Table 15. Although the data in Table 15 are specific and are presented only for illustration, data on the isotopic composition of irradiated plutonium, available from other sources,^(17,48) permit similar calculations to be made for any fuel.

Table 15

CALCULATED DOSE RATES AND ISOTOPIC COMPOSITION OF PLUTONIUM FROM A POWER REACTOR FUEL⁽⁴⁰⁾

Dose Rates From a 1-kg Sphere of Plutonium (rad/hr)

Isotope	Exposure - 1000 MWD/T						Exposure - 3000 MWD/T						
	Conc. (%)	Time After Separation (yr)				n/sec-kg Pu ^a	Conc. (%)	Time After Separation (yr)				n/sec-kg Pu ^a	
		0	0.1	0.2	1.0			0	0.1	0.2	1.0		
Pu ²³⁸	0.002	0.015	0.015	0.015	0.015	4.7 x 10 ⁴	0.02	0.15	0.15	0.15	0.15	4.7 x 10 ⁵	
Pu ²³⁹	90.2	0.97	0.97	0.97	0.97	4.0 x 10 ⁶	73.5	0.79	0.79	0.79	0.79	3.3 x 10 ⁶	
Pu ²⁴⁰	9.50	0.62	0.62	0.62	0.62	1.4 x 10 ⁶	19.8	1.44	1.44	1.44	1.44	3.3 x 10 ⁶	
Pu ²⁴¹	1.27	0.16	0.16	0.16	0.16	2.3 x 10 ³	6.04	0.75	0.75	0.75	0.75	1.07 x 10 ⁴	
Pu ²⁴²	0.028	-	-	-	-	6 x 10 ¹	0.64	-	-	-	-	1.4 x 10 ³	
U ²³⁷	-	-	0.60	0.60	0.60	-	-	-	2.68	2.68	2.68	-	
Am ²⁴¹	-	-	0.19	0.35	1.78	7.4 x 10 ⁴ (0.1 yr)	-	-	0.86	1.69	8.43	3.1 x 10 ⁵ (0.1 yr)	
Total β, γ	-	1.765	2.56	2.72	4.15	-	-	3.13	6.85	7.68	14.42	-	
Total n (S.F.) ^b	1.2 x 10 ⁵							2.95 x 10 ⁵					
Total n (α,n)						5.5 x 10 ⁶							7.4 x 10 ⁶

^aNeutrons from 1.32 kg PuF₄ due to (α, n) reactions. The Am²⁴¹ value calculated for 0.1 yr after separation. These data do not include neutron multiplication.

^bS.F. = Spontaneous Fission.

In summary, the primary hazard from handling plutonium, particularly small quantities, is an internal one, as evidenced by the very low values for the maximum permissible body burden. Consequently, the special enclosures used for work with plutonium serve primarily to confine the material and reduce the inhalation hazard. When larger quantities of plutonium are involved, exposure from gamma and X rays as well as from fast neutrons becomes important. The contamination of plutonium by americium-241 contributes significantly to the dose rate from highly irradiated material.

V. AMERICIUM

Although recent emphasis on the application of nuclear energy to thermoelectric devices (SNAP Program) has increased interest in americium, the quantities of purified americium available for research and development are relatively small. Americium-241, obtained from aged plutonium containing appreciable quantities of plutonium-241, is perhaps the most important isotope of this element. Irradiation of americium-241 leads to the formation of highly radioactive curium-242 and plutonium-238. In addition, irradiation of americium-241 (or plutonium containing appreciable quantities of americium-241) leads to the formation of americium-244, which is the precursor of most of the transamericium isotopes. The decay constants for several isotopes of americium are shown in Table 16.

Table 16

DECAY CONSTANTS OF SELECTED AMERICIUM ISOTOPES⁽³⁰⁾

Isotope	Half-life	Specific Activity (d/sec-g)	Decay Scheme ^a
Am ²⁴¹	458 yr	1.2×10^{11}	α (5.477, 5.435, 5.378); γ (26, 59.6, 99, 103)
Am ²⁴²	16.01 hr	2.99×10^{16}	β^- (81%); E.C.* (19%); γ (42, 44)
Am ^{242m} ^{b,c}	152 yr	3.41×10^{11}	β^- (83.6%); E.C. (18.9%); α (0.476%)
Am ²⁴³	7950 yr	6.85×10^9	α (5.266, 5.224, 5.169); γ (75)
Am ²⁴⁴	26 min	1.1×10^{18}	β^- (99+%); E.C. (0.039%)

^aValues in parentheses are energies (Mev for α , kev for γ) of the principal particles or quanta, or the percent of the decays proceeding by the mode indicated.

^bBarnes, R. F., *et al.*, J. Inorg. Nucl. Chem., 9, 105 (1959).

^cAsaro, F., and Perlman, I., Phys. Rev., 120, 934 (1960).

*E.C. = electron capture.

Little experimental data are available on the external dose from americium. In a discussion of the conversion of americium oxalate to the oxide, Baybarz⁽⁴⁹⁾ indicated that a 30-gm batch of americium dioxide in a glass bottle showed a reading of ~12 r/hr with a "Cutie Pie" survey meter.**

**This instrument has a 1-mg/cm² Mylar window.

Lead-impregnated neoprene gloves decreased the radiation to approximately 150 mr/hr. A one-gram sample of americium-241 used as a calibration standard for film badges shows a dose rate of 230 mr/hr at 20 cm.⁽⁵⁰⁾ Examination of the decay scheme of americium-241 shows that the 60-kev gamma ray, representing approximately 40% of the disintegrations, is the major contributor to the external hazard.

The neutron hazard from larger samples of americium may be appreciable even if such low yield elements as oxygen are considered. For example, in connection with the handling of americium dioxide, a surface fast neutron dose rate of 740 mr/hr has been calculated⁽⁵¹⁾ for a 100-gm sample of $\text{Am}^{241}\text{O}_2$ contained in a glass vessel, 6 cm in diameter and 3.5 cm high. Although no data on the method of calculation are given, this value is probably much too low if the neutron energy is assumed to be such that $1 \text{ nf/cm}^2\text{-sec} = 0.125 \text{ mr/hr}.$ *

The internal hazard from americium-241 is similar to that from plutonium,⁽⁹⁾ the critical organs being the bone and kidney for soluble americium and the lung and GI tract for insoluble americium. The maximum permissible body burden for soluble americium-241 is limited to $0.05 \mu\text{c}$ by the dose to bone tissue.

*On the assumption that the yield integral is 0.19×10^{-6} , a 100-gm sample at unit density has a calculated surface flux of $3.3 \times 10^6 \text{ n/cm}^2\text{-sec}.$

VI. PROTACTINIUM, NEPTUNIUM, AND THE TRANSAMERICIUM ELEMENTS

A. Protactinium

Protactinium is found in nature as a result of the decay of uranium-235 (protactinium-231) and uranium-238 (protactinium-234). Protactinium-234 is a short-lived beta emitter which decays to uranium-234. Protactinium-231 is a long-lived alpha emitter found in pitchblende ore at a concentration of approximately 0.1 ppm. The very low concentration of protactinium* has made isolation of larger quantities of this material uneconomical, but recently⁽⁵²⁾ 125 gm of protactinium were isolated and purified. The internal hazard from protactinium-231 is comparable with that from natural thorium.⁽⁹⁾ No data have been found which evaluate quantitatively the external hazard, but the decay of protactinium-231 is accompanied by a variety of gamma rays.

Both protactinium-233 and -234 are produced by neutron irradiation of thorium-232. The relatively large amounts of protactinium-233 present in irradiated thorium contribute a major portion of the radiation hazard from fission-product-free thorium recently discharged from reactors. Both of these isotopes decay by beta and gamma emission with relatively short half-lives to the corresponding uranium compound, and the external hazard from these isotopes appears to be paramount even when relatively small quantities are involved. Since protactinium-233 is usually either allowed to decay or is separated from thorium and uranium, the radiations from these isotopes are of little interest at this point.

B. Neptunium

Two important isotopes of neptunium are produced by neutron irradiation of uranium. Neptunium-239, resulting from the decay of uranium-239, forms the parent substance for plutonium-239. Although neptunium-239 has a short half-life, its concentration in discharged power reactor fuel may be relatively high. Neptunium-237, formed from the beta decay of uranium-237, is a long-lived alpha emitter, with a half-life of 2.2×10^6 yr. The internal hazard from neptunium-239 is less than that for neptunium-237, and this is reflected in the values for the maximum permissible body burden, which are 30 μ c (bone) and 0.06 μ c, respectively. The latter indicates an internal hazard comparable to that for plutonium. Although relatively large quantities of neptunium-237 have been produced,⁽⁵³⁾ no data have been found which permit quantitative evaluation of the external hazard from neptunium, but the long half-life of neptunium-237 points toward relatively small gamma hazard.

*Natural uranium contains only 0.34 ppm protactinium-231 at equilibrium.

C. Transamericium Elements

All of these elements are formed by neutron irradiation of plutonium or americium. Curium isotopes of mass numbers 240 to 249 have been isolated and identified. Irradiation of americium-241 yields curium-242, which is utilized together with its decay product, plutonium-238, in thermoelectric devices employing radioactive decay as the source of heat. Irradiation of curium-249 yields berkelium-249 and -250 which, in turn, are the precursors for the californium series of mass numbers 249 to 254. Although the quantities of these elements which have been produced are generally extremely small, the short half-lives and intense radiation contribute to a significant external hazard. Further, several isotopes of curium, berkelium, and californium have very short spontaneous fission half-lives and thus emit a large number of fast neutrons. In several instances, the neutron flux from submicroscopic samples of material represents a serious external hazard.

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VII. SUMMARY

A survey of the hazards connected with the handling of actinide elements shown in Table 17 has been presented to acquaint the reader with the scope of the problems likely to be encountered. The chemical toxicity of the actinides is usually minor when compared with the radiological toxicity and therefore has not been treated in this discussion. The values of maximum permissible body burden expressed in microcuries, when used as a measure of the relative hazards, show the actinides to be rather comparable in toxicity. Thus, inhalation and ingestion are the important routes of entry; direct injection into the blood stream through wounds also requires consideration. Special enclosures, such as glove boxes, function primarily to minimize the risk of inhalation and to aid in confinement.

Table 17

SUMMARY OF HAZARDS FROM THE ACTINIDE ELEMENTS

Element	Internal	External			
		Beta	Gamma	Neutrons	
				(α, n)	Spontaneous Fission
Thorium	0.01 μc (natural) 9 x 10 ⁴ μg Th ²³²	Surface dose 107 mrem/h from natural thorium, in- creases with Th ²²⁸ content.	Surface doses 85 mrem/h from natural thorium, in- creases with Th ²²⁸ content.	Negligible	Negligible
Uranium	0.005 μc (natural U) 1.4 x 10 ⁴ μg natural U 0.84 μg U ²³³	Surface dose 240 mrep/h from natural U (beta- gamma). In- creases with U ²³² content.	Increases sharply with U ²³² content.	High for uranium containing U ²³² .	Small (negligible for natural U).
Neptunium	0.06 μc (Np ²³⁷) (71 μg Np ²³⁷)	Negligible	Small	High	Negligible
Plutonium	0.008 μc (Pu ²³⁹) (0.13 μg Pu ²³⁹) (0.0024 μg Pu ²³⁸) (0.0082 μg Pu ²⁴⁰)	Nil except for Pu ²⁴¹	Medium to high depend- ing on the isotopic composition.	High	Medium to high depend- ing on the isotopic composition.
Curium, Berkelium Californium, etc.	0.04 μc (minimum)		High	High	Very high for certain isotopes.

The external hazard from actinide elements, primarily due to gamma-ray and fast neutron emission, varies considerably with the element and its source. Irradiated actinides, such as thorium and plutonium, usually show an increase in the external hazard from gamma radiation with extent of irradiation. As is evident in the case of plutonium, the time after separation from impurities and the degree of decontamination accomplished strongly influences the gamma dose. Finally, fast neutrons from (α, n) reactions and spontaneous fission decay represent a difficult problem in many instances, particularly in the terminal elements of the actinide series.

No consideration has been given to several topics which may simply be mentioned to increase awareness of their severity. The transuranic elements as well as uranium-235 contain fissile isotopes, and thus criticality considerations are superimposed on the handling hazards mentioned in this review.⁽⁵⁴⁾ Secondly, while neutron fluxes from natural uranium owing to spontaneous fission or (α, n) reactions are very small, neutron irradiation of even small samples of uranium in such facilities as "rabbits" may produce large fluxes from delayed neutrons. The magnitudes of these fluxes clearly vary with irradiation time and the delay in removing the samples from the reactor, but close attention to the delayed neutron flux is normally required when very short decay times are used.⁽⁵⁵⁾ Finally, concentrated aqueous solutions of actinide elements will evolve hydrogen and oxygen owing to the radiolytic decomposition of water. The rate of gas evolution is strongly dependent on the solutes, but in the multigram-per-liter concentration range this phenomenon deserves attention, particularly when storing solutions in closed containers.

Although considerable data have been presented which may permit calculation of the external hazards of a particular situation, it is necessary to emphasize that measurement of the radiation field affords a more reliable evaluation. Dose measurements to the hand with its attending higher permissible exposure may, for example, represent a better evaluation of the hazard than estimation of the total body exposure by calculation. The magnitude of the neutron flux will usually be dependent on the surroundings of the source, owing to scattering, moderation, and absorption. Thus, while the topics treated here may serve to generate awareness, measurements are necessary to define quantitatively the problems in the light of the exposure limits currently in use.

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