Safety Reports Series

No.9

SAFE HANDLING AND STORAGE OF PLUTONIUM

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FOREWORD

A large increase in the use of plutonium was anticipated with the development of the civil applications of nuclear energy. Significant capital investments were made in facilities to separate plutonium from spent nuclear fuel. However, with the discoveries of large quantities of inexpensive uranium ores (which provide an alternative to plutonium as a nuclear fuel) coupled with the slow development of nuclear electrical energy and with the rapidly escalating cost of developing and deploying fast breeder reactors (which were expected to be the major users of plutonium), the utilization of separated plutonium has not kept pace with its rate of separation. As a result, the stockpiles of separated civil plutonium around the world totalled more than 150 t at the end of 1996.

This Safety Report updates IAEA Safety Series No. 39, ‘Safe Handling of Plutonium’, which was published in 1974. The focus of the previous publication was on plutonium research and development facilities, which used very limited quantities of plutonium. At that time, the average burnup of fuel was much lower than it is today. With higher burnup, there are higher concentrations of ^{238}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu. Also, large amounts of weapon grade plutonium (with greater than 90% ^{239}Pu) have been declared to be in excess of military requirements, and these materials may also be added to the civil plutonium inventories. This report therefore describes the effects of this wide variance in isotopic composition on storage and handling requirements. The effects of stricter standards for the exposure of personnel to radiation — which have been established since Safety Series No. 39 was published — are also described. That publication did not address criticality because it covered only laboratory scale facilities (facilities which had less than 220 g of plutonium). This report, however, describes facilities now in place or needed in the future which have large quantities of plutonium, and thus it addresses criticality issues as well. Further, because of the growing need to store plutonium for long periods of time, this report also covers plutonium storage. Although safeguards and physical security are very important issues with respect to plutonium handling and storage, these issues are not covered in this report.

The development and publication of this report is part of an expanded programme within the IAEA to identify and deal with problems associated with the accumulation of stockpiles of separated civil plutonium. It is the result of a sharing of data and experience concerning the handling and storage of plutonium by the countries which have the most experience in these areas. The officer responsible for this report was J. Finucane from the Nuclear Fuel Cycle and Materials Section of the Division of Nuclear Fuel Cycle and Waste Technology.

CONTENTS

1. INTRODUCTION

1.1. BACKGROUND

In the early 1970s, demand for electricity was growing at more than 7% per year, leading to a doubling of demand every ten years. It was anticipated that nuclear power would supply most of the new electricity generating capacity. After the oil crisis of 1973, the growth in electricity demand dropped to 3% per year, but the shortage of oil strengthened the option for nuclear capacity in countries dependent on oil imports. The availability of uranium was a major concern at that time.

Early nuclear power activities were focused on the development and deployment of light water reactors (LWRs) and gas cooled reactors (GCRs) and on the development of fast reactors, which were considered to be the most efficient systems for resource utilization. In fact, the first power reactor was a fast reactor (EBR-1). The initial inventory of plutonium was obtained by reprocessing fuel from thermal reactors.

With the discovery of large uranium deposits, primarily in Canada and Australia, uranium prices dropped considerably, weakening the justification for fast reactor systems. Further, with the escalation in capital costs associated with enhanced safety requirements and the reduction in fossil fuel prices, the large scale deployment of fast reactors has been delayed and is not expected to commence before 2030.

Mixed oxide fuel (MOX) for thermal reactors uses fissile plutonium in place of some ^{235}U. The fuel is fabricated in several plants around the world, most commonly by blending plutonium oxide and uranium oxide. The fabrication of MOX is similar to the fabrication of uranium oxide fuel. Although the use of MOX fuel in thermal reactors had been demonstrated for more than 30 years, plutonium was separated and stockpiled primarily for use in fast reactors. However, with the reprocessing capacity already installed, and the production of tonne quantities of plutonium annually, this resource is now being utilized to provide the fissile content of thermal reactor MOX fuel on an industrial scale. By 1995, over 300 t of MOX fuel containing 15 t of plutonium had been used to fuel LWRs.

Over the past 25 years (since the IAEA’s Safety Series No. 39 [1] was published), power reactors have generally moved to higher and higher average burnups, which result in higher concentrations of ^{238}Pu, ^{240}Pu, ^{241}Pu and ^{242}Pu in the spent fuel. The effects of the increases in the concentration of these isotopes are described in this report.

With the end of the Cold War and a reduction in the stockpiles of nuclear weapons, about 50 t of Russian and US plutonium may be designated as no longer being required for defence purposes. The USA has declared that 52.7 t are excess to its defence needs and has announced a dual-track approach for eliminating its excess weapon plutonium — using some of the plutonium in MOX fuel in a limited number of existing US power reactors, and immobilizing some of the plutonium in glass or ceramic waste form, along with high level wastes, for disposal. The Russian Federation is continuing its studies. The potential need to handle and store plutonium that is designated as no longer being required for defence purposes is also described in this report.

In 1995, there was installed reprocessing capacity of about 2590 tonnes per annum (t/a) for water reactor fuel and 2100 t/a for GCR fuel, with an additional 2100 t/a LWR reprocessing capacity at various stages of planning or development. Over the next 15 years, GCR fuel reprocessing capacity of about 1300 t/a will be retired. In 1996, there was about 100 t/a of MOX fabrication capacity installed, with over 200 t/a under construction and a further 200 t/a capacity being planned.

Commercial scale handling of plutonium involves compliance with stringent safety requirements, and this report addresses these issues as well. It should be noted that the safety standards that applied to personnel exposure in the early 1970s (derived from International Commission on Radiological Protection (ICRP) publications [2–4]) have become more stringent to comply with ICRP Publication 60 (1991) [5]. The IAEA has published the International Basic Safety Standards (1996) [6] containing the latest safety recommendations.

1.2. OBJECTIVE

The objective of this safety report is to describe in a single publication the best current practices for handling and storing plutonium while providing concrete examples of the safe design and operation of commercial scale facilities. These practices are derived from widely accepted radiation protection and safety principles such as those published by the ICRP and in IAEA Safety Series publications.

1.3. SCOPE

This publication updates the earlier Safety Series No. 39, ‘Safe Handling of Plutonium’. While that publication was concerned only with laboratory scale operations and thus did not include criticality, this safety report deals with industrial scale operations and therefore does include criticality. It also includes the effects of a broader range of isotopic concentrations and of the more restrictive radiation exposure standards. The intention is to provide not only the practice but also to describe the approaches to nuclear safety developed over many years of industrial scale activities.

This report is intended to act as a concise source of information for those who need to understand the basis for the safe handling and storage of plutonium, and as such contains supporting information on the effects and behaviour of plutonium. It does not contain detailed information on procedures, as these are developed by operating plants. Although safeguards, physical protection and export controls for plutonium are very important, they are not covered here; the transportation of plutonium is also not covered as this topic is covered in other IAEA publications.

1.4. STRUCTURE

The scope of plutonium operations in the mid-1990s and projections to 2010 are provided in Section 2. The nuclear, physical and chemical properties of the different forms of plutonium are briefly reviewed in Section 3. Environmental behaviour and monitoring are described in Section 4. Pathways to humans and the biological behaviour of plutonium are described in Section 5. The constraints provided by regulatory limits are outlined in Section 6. Section 7 provides the basis of best current practice in the design of a modern commercial scale plant, while Section 8 describes some of the general operating procedures in the safe operation of a plutonium processing plant. Section 9 provides a brief summary of this report.

Supplementary reference material is contained in the annexes. Annex I contains examples of the design and operation of different types of plants, while Annex II contains radiological safety data for handling plutonium. Annex III contains tables of criticality parameters. A glossary of terms and acronyms which may not be familiar to readers of this report is also included.

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2. CURRENT AND FUTURE PLUTONIUM ACTIVITIES AND INVENTORIES

Activities concerning the separation and use of plutonium can conveniently be subdivided into separation, conversion, storage, fuel fabrication and irradiation (and perhaps repetitions of these steps). The information provided in this section describes the status in 1997 and projections of reprocessing and MOX fabrication capacities until 2015. These projections are not intended to predict the future circumstances, but only to indicate the scope of expected future activities.

2.1. SEPARATION OF CIVIL PLUTONIUM FROM IRRADIATED FUEL

Separated plutonium is obtained by reprocessing irradiated nuclear fuel. Table I gives estimates of the capacities of reprocessing facilities around the world. The amount of plutonium separated depends on both facility throughput and the plutonium content of the spent fuel. The throughput of any facility depends on the ‘demand’ for reprocessing and may be considerably less than capacity. The plutonium content of the spent fuel, which depends primarily on burnup of the fuel, is typically 6–12 g/kg of uranium for LWR fuel (~1%) and 2–3 g/kg of uranium (~0.3%) for GCR fuel. Reprocessing is efficient, with greater than 99.5% of plutonium being recovered [1].

The Russian Federation and the USA have announced that they have plutonium in excess of their military needs and may transfer it to civil status for use in reactors or dispose of it as waste immobilized in a geological repository. However, the timing of the transfer has not been determined. Eventually, the other nuclear weapon States may also declare some quantity of plutonium as being in excess of military needs.

2.2. STORAGE OF SEPARATED CIVIL PLUTONIUM

At the end of 1995, about 47 t of separated civil plutonium were stored at Sellafield (British Nuclear Fuels Limited (BNFL)) in the United Kingdom, about 55 t in France, about 27 t at Production Association MAYAK in the Russian Federation, and about 5 t in Japan. These figures include nearly all of the worldwide inventory of separated civil plutonium (Fig. 1). This inventory does not include any excess military material.

TABLE I. SUMMARY OF REPROCESSING CAPACITY[a] (t HEAVY METAL)

	1997	2000	2005	2010	2015
LWR					
United Kingdom (BNFL Thorp)	600	600	600	600	800
France (Cogéma UP2)	800	800	800	800	800
France (Cogéma UP3)	800	800	800	800	800
Japan (Tokai)	90	90	90	90	90
Japan (Rokkashomura)			500	800	800
Russian Federation (RT-1)	400	400	400	400	400
Russian Federation (RT-2)[b]			(500)	(1000)	(1000)
Total LWR	2690	2690	3190	3490	3490
GCR					
United Kingdom (BNFL-THORP-AGR)	200	200	200	200	200
United Kingdom (BNFL-Magnox)	1500	1500	1500	0	0
France (Cogéma-UP1)	430				
Total GCR	2130	1700	1700	200	200
PHWR					
India	200	600	600	600	600

[a] Throughput depends on ‘demand’ and therefore may be significantly less than capacity.

[b] Linked to RT-2 plant status, not included in total.

2.3. CONSUMPTION OF SEPARATED CIVIL PLUTONIUM

At present there are large inventories of separated plutonium and more plutonium is being separated than is being used. Currently the primary use of separated civil plutonium is as MOX fuel in LWRs (see Fig. 1). Some plutonium is also being used in fast reactor R&D work. A complementary way to use plutonium appears to be in reconfigured fast reactors, which may then consume large quantities of plutonium. In some countries plutonium may eventually be disposed of as radioactive waste.

2.3.1. Use of MOX fuel in LWRs

The reuse of plutonium in power reactors as MOX fuel is a major step to reduce the inventory of plutonium produced in conventional LWRs while utilizing the energy

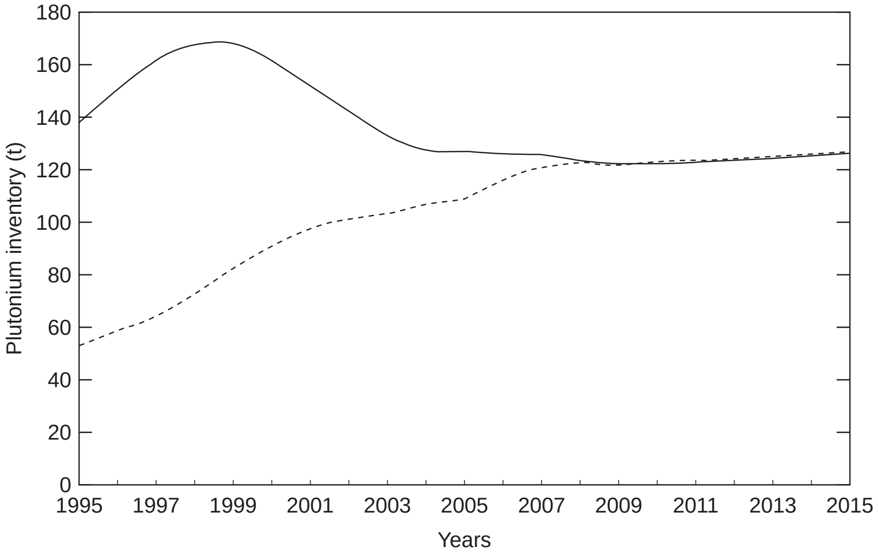

FIG. 1. Projected worldwide separated plutonium and minimum plutonium inventories (—— : end of year plutonium inventory; ----- : minimum plutonium inventory).

TABLE. II. MOX FABRICATION CAPACITIES (t HEAVY METAL)

	1997	2000	2005	2010	2015
Belgium (Dessel) PO	40	40	40	40	40
France (CFCa)	30	40	40	40	40
France (Melox)	90	210	250	250	250
India[a]	(20)	(20)	(20)	(20)	(20)
Japan (Tokai)	13	13	13	13	13
Japan-LWR MOX			100	100	100
Russian Federation[b]				(150?)	(150?)
United Kingdom (MDF)	8	0	0	0	0
United Kingdom (SMP)		100	120	120	120
Total	181	403	563	563	563

[a] Throughput is minimal to match demand, not included in total.
[b] Linked to RT-2 plant status, not included in total.

produced. As a result, the MOX fabrication plants tend to be operated at their indicated capacities (Table II) and the throughput is equal to the capacity (except as noted).

A typical LWR fuel contains 6–12 kg/t heavy metal (HM) of plutonium after irradiation (1%). Typical MOX fuel for PWRs contains about 60 kg/t HM of plutonium before irradiation and only about 40 kg/t HM of plutonium after irradiation. Thus, burning MOX does consume plutonium. Thirty years of experience exists in using MOX fuel in LWRs; a total of 300 t of MOX fuel, containing 15 t of plutonium has already been loaded in LWRs. At present there are more reactors licensed to burn MOX than there is MOX fuel available; the French utility Electricité de France plans to load MOX fuel into 28 LWRs before the year 2000. Stocks of separated plutonium will depend on its production by reprocessing and on the capacity of MOX fabrication plants. Analyses have been conducted which conclude that the separated plutonium inventory will stabilize and then decrease at the beginning of the next century. The time and amount of the maximum inventory will depend on the reprocessing and MOX fabrication throughput rates [2]. Consumption of about 8 t of plutonium annually in the mid-1990s is expected to increase to about 34 t annually by 2010.

2.3.2. Use of plutonium fuel in fast reactors

The consumption of plutonium in fast reactors depends upon the extent of the deployment of such reactors, which in turn depends upon energy prices, successful R&D, public acceptability, uranium availability and cost, and reduction in the capital cost of fast reactors. In the near term, only limited use of plutonium in fast reactor R&D is anticipated.

There is now a broad consensus that fast reactors can be used as plutonium burners. Significant fast reactor technology development programmes are proceeding in France, the Russian Federation, Japan and, to a lesser extent, in India, the Republic of Korea and China. Some of the programmes are aimed at using the fast reactor for utilizing plutonium stocks and burning minor actinides. In 1993, the French launched a programme to adapt the fast reactor to consume plutonium and thus help reduce the plutonium inventory [3, 4]. The proposed fast reactor will use a mixture of plutonium oxide and uranium oxide as fuel. In terms of fabrication, use and reprocessing, it is similar to the MOX fuels already in use in LWRs. However, it will use a higher fraction of plutonium in the fuel and will therefore consume more plutonium. With fuel that contains 45% plutonium (compared with 5–7% plutonium in LWR MOX fuel), the net plutonium consumption is about 70 kg/TW·h(e) (terawatt hour of electricity produced), compared with no net consumption for LWRs loaded at 30% with MOX fuel and with a consumption of 60 kg/TW·h(e) for LWRs loaded at 100% with

MOX fuel. A further step in the effort to reduce plutonium inventories could be a uranium free fast reactor fuel — for which plutonium consumption could reach as high as 110 kg/TW·h(e) produced.

2.3.3. Disposal of plutonium as waste

The USA announced in early 1997 that one of its preferred alternatives is to dispose of some of its plutonium designated as no longer required for defence purposes as waste immobilized in either a glass or ceramic waste form. However, the details of this option are still being evaluated and the timing of such an operation has not yet been determined.

2.4. RESEARCH AND DEVELOPMENT: EMERGING TECHNOLOGIES

Various R&D efforts are being conducted to develop new technologies with the aim of reducing the inventories of separated civil and excess military plutonium and the waste arising from plutonium reprocessing.

2.4.1. Long term strategies for reducing plutonium inventories

To define the strategies for the long term use of plutonium, current R&D is concentrating on the following to:

- Increase MOX content in present reactor cores above the current 30%,
- Develop multirecycling of plutonium,
- Design a new reactor capable of handling 100% loading of MOX,
- Investigate using fast reactors as plutonium burners,
- Create a stable form for geological disposal.

2.4.2. Waste reduction

There are R&D efforts under way to:

- Optimize existing processes to reduce the amount of waste.
- Recycle these wastes in the process [5].
- Decontaminate to allow disposal as low level waste. A typical example is the recent development of a specific electrochemical dissolution process which efficiently decontaminates miscellaneous wastes, such as ashes, from incineration or plutonium contaminated metallic surfaces [6].
- Study the feasibility of removing minor actinides.

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3. NUCLEAR, PHYSICAL AND CHEMICAL PROPERTIES OF PLUTONIUM

3.1. NUCLEAR PROPERTIES OF PLUTONIUM

Plutonium is produced in reactors as a result of initial neutron capture by ^{238}U to produce ^{239}U, which beta decays first to ^{239}Np and then to ^{239}Pu, as shown in Fig. 2 [1]. The ^{240}Pu, ^{241}Pu, ^{242}Pu and ^{243}Pu isotopes are formed by successive neutron capture. Most of the ^{238}Pu is also formed by (*n*, *2n*) reactions. From a practical perspective, isotopic production terminates at ^{242}Pu because the half-life of ^{243}Pu is only 5 h.

Plutonium can be produced in reactors with any type of neutron spectrum — thermal, intermediate or fast. The isotopic composition of the plutonium varies depending on the burnup of the fuel at discharge and on the level of uranium enrichment. Examples of isotopic distributions for different burnup levels and reactors are given in Table III. As can be seen, the isotopic composition is strongly dependent on the extent of burnup.

As shown by data in Table IV, the nuclear properties of the plutonium isotopes differ [2]. As a result, the nuclear characteristics of plutonium are strongly dependent on the isotopic compositions. Note particularly that ^{241}Pu (half-life of 14.4 a) beta decays to ^{241}Am, which alpha decays with emission of a 59 keV gamma to the ^{237}Np daughter. Consequently, the gamma radioactivity resulting from ^{241}Am is larger for plutonium from high burnup fuel than from low burnup fuel and it increases over time to a maximum that is reached after 72 a. The fraction of each plutonium isotope remaining as a function of time is shown for a one hundred year period in Fig. 3. A curve showing the ^{241}Am fraction relative to the initial amount of ^{241}Pu present is also included.

Criticality is an important nuclear consideration in handling plutonium. The radiation level during a criticality event is extremely high and may be fatal to personnel in the immediate vicinity. The ^{239}Pu and ^{241}Pu isotopes have relatively high fission cross-sections compared with ^{240}Pu and ^{242}Pu [2]. The critical mass of an unmoderated and unreflected sphere of alpha-phase ^{239}Pu (19.8 g/cm^3) is about 10 kg, whereas the water reflected value is 5.6 kg [1]. For a given mass of plutonium, the specific reactivity decreases as the percentages of ^{240}Pu and ^{242}Pu increase. The critical mass for reactor grade plutonium is about twice that of ^{239}Pu. Additional criticality concerns exist for plutonium solutions owing to increased neutron moderation and reflection in solvents with a large hydrogen content. The minimum critical mass in a water moderated and reflected spherical geometry is approximately 0.5 kg of ^{239}Pu at a concentration of about 7 g/L.

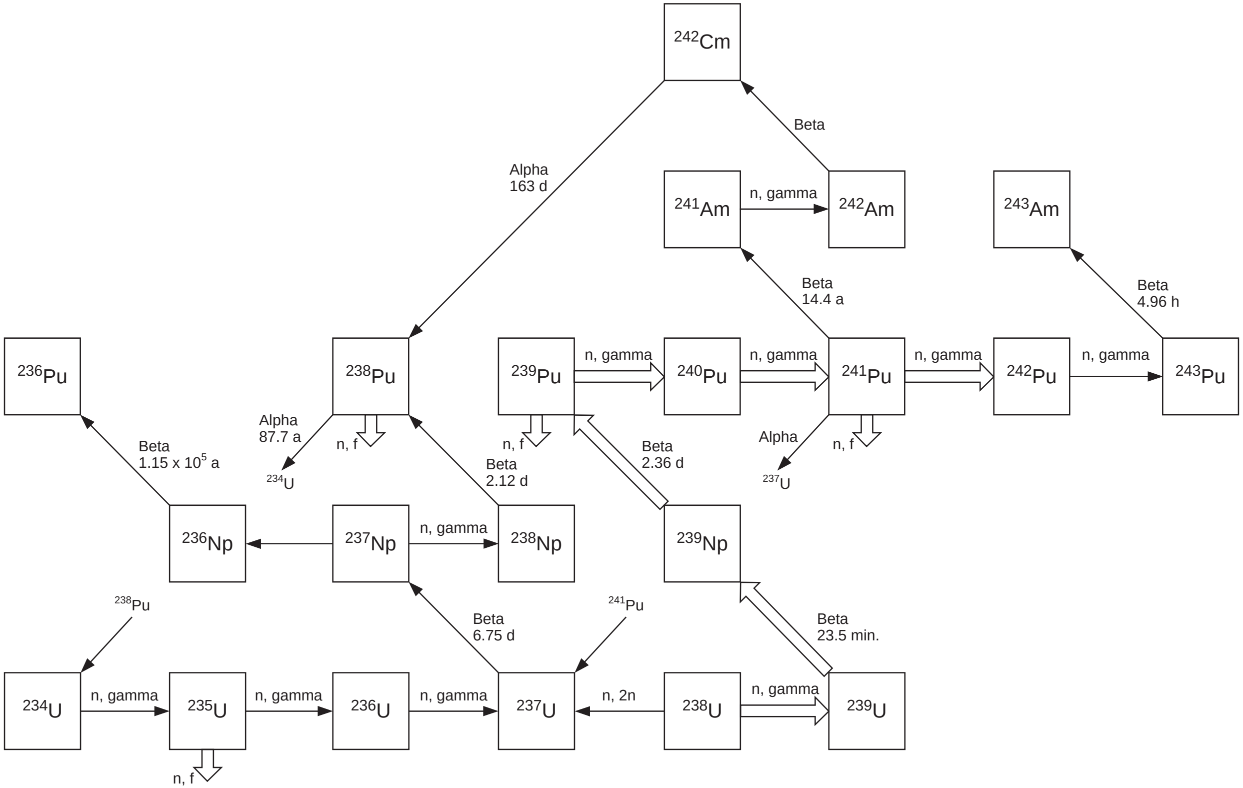

FIG. 2. Principal modes of plutonium production through the neutron irradiation of uranium (⇒: principal modes of production; →: normal radioactive decay; ⇓: fission).

TABLE III. EXAMPLES OF TYPICAL PLUTONIUM ISOTOPIC PERCENTAGES FOR DIFFERENT BURNUP LEVELS AND REACTOR DESIGNS

Material type	Reactor type	Burnup (GW·d/t)	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu[a]	^{242}Pu
Production grade ^{238}Pu	—		80.0	16.3	3.0	0.6	0.1
Medical grade ^{238}Pu	—		90.4	9.0	0.6	0.3	0.1
Weapons grade Pu	Production	<1	0.04	93.3	6.0	0.6	0.04
Spent fuel	MAGNOX	5	—[b]	68.5	25.0	5.3	1.2
Spent fuel	CANDU	7.5	—[b]	66.5	26.5	5.3	1.5
Spent fuel	LWR	20	0.5	73.5	20.0	5.0	1.0
Spent fuel	LWR	30	1	60	22	13	4
Spent fuel	LWR	60	4.4	46.3	24.9	12.7	11.7

[a] The amount of ^{241}Am depends on the initial ^{241}Pu content and time since reprocessing (see Fig. 2 for data to determine the ^{241}Am content).

[b] ^{238}Pu is formed, but the percentage is very small.

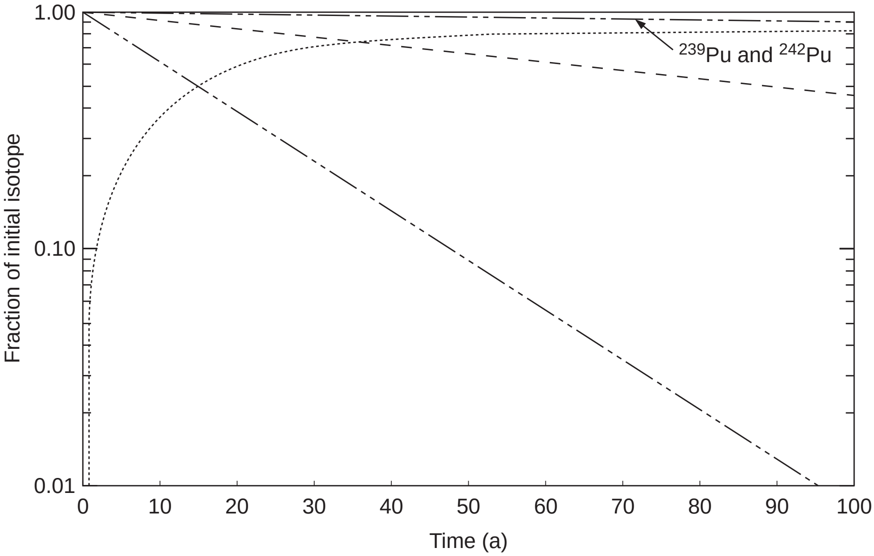

FIG. 3. Fraction of plutonium isotopes remaining after separation and buildup of ^{241}*Am (– – –:* ^{238}*Pu; – - - - –:* ^{240}*Pu; -------:* ^{241}*Am/*^{241}*Pu initial).*

External radiation hazards exist for personnel during the handling of plutonium because the radioactive decay of commonly encountered plutonium isotopes produces gamma rays and X rays, and ^{240}Pu and ^{242}Pu produce spontaneous neutrons (see Tables IV and V). Low and medium energy gamma rays, mainly from ^{241}Pu decay products (primarily ^{241}Am), are the major contributors to the external radiation exposure of personnel working with power reactor origin plutonium. Therefore, special attention is given to the ingrowth of ^{241}Am. The gamma emissions from other plutonium decay products may become the major external gamma radiation source if the ^{241}Am is either removed or the low energy gamma rays are attenuated by shielding. Additional neutron radiation arises from (α, n) reactions that occur when light nuclei (e.g. Be, O, F and Al) are bombarded by alpha particles [3]. The neutrons have a wide energy distribution with a maximum of 13 MeV and mean energies of 1–5 MeV, depending on the nuclei involved. The energies of the alpha particles from the decay of plutonium nuclei and ^{241}Am range from 4.9 to 5.5 MeV. X rays with energies of 12–17 keV are emitted by uranium daughters, but occur in significant percentages of the decay events only for ^{238}Pu (13%), ^{240}Pu (50%) and ^{242}Pu (10%).

Radiation damage to tissues, which is a potential consequence of plutonium exposure during handling and storage, results primarily from the alpha activity of plutonium deposited in the body by inhalation, ingestion or wounds. Alpha emission

TABLE IV. NUCLEAR PROPERTIES OF PLUTONIUM AND RELATED ISOTOPES

Isotope	Half-life (a)	Decay mode[a]	Specific activity (10^9 Bq/g)	Spontaneous neutron rate (n/g per s)	Heat generation (W/kg)	He gas production rate (mmol/kg per year)	Product isotope
^{236}Pu	2.85	α	18 600	3.7×10^4	17 900	1000	^{232}U
^{238}Pu	87.7	α	600	2.62×10^3	560	32.6	^{234}U
^{239}Pu	2.41×10^4	α	2	0.03	1.9	0.12	^{235}U
^{240}Pu	6.54×10^3	α	8	1.02×10^3	6.8	0.44	^{236}U
^{241}Pu	14.4	β[b]	3700	—	4.2	—	^{241}Am
^{241}Pu	7.2×10^5	α[b]	0.07	8.8×10^2	0.06	0.004	^{237}U
^{242}Pu	3.76×10^5	α	0.1	1.7×10^3	0.1	0.01	^{238}U
^{241}Am	4.32×10^2	α	120	1.1	114	6.8	^{237}Np

[a] All decay processes are accompanied by the emission of some X rays and gamma rays.

[b] Approximately 0.002% of ^{241}Pu decays by alpha particle emission to ^{237}U.

TABLE V. SURFACE DOSE RATES FOR 1 kg SPHERES OF PURE NUCLIDE

Isotope	X rays (mSv/h)	Gamma rays (mSv/h)	Spontaneous neutrons[a] (mSv/h)
^{238}Pu	5700	240	640
^{239}Pu	89	3.2	<0.01
^{240}Pu	72	0.8	300
^{241}Pu	—	120	—
^{242}Pu	1.3	—	310
^{241}Am	4000	27 000	0.15

[a] Neutron rates are from spontaneous fission processes; contributions from (α, n) reactions must be added for oxides and other compounds of plutonium with light elements (carbon, nitrogen, etc.).

is a major process in the decay schemes of all major plutonium isotopes and/or their daughter radionuclides. Therefore, containment of plutonium is essential. The ^{238}Pu isotope is a major concern because of its high specific activity (see Table IV). The ^{238}Pu content increases with increasing fuel irradiation and exceeds 2% for burnup levels of 40 GW·d/t in LWR reactors.

A final concern is heat generation (self-heating) from radioactive decay during storage and handling. Whereas ^{239}Pu generates about 1.9 W/kg, the thermal output of ^{238}Pu is approximately 560 W/kg. Owing to the large content of ^{239}Pu in civil and military materials and to the high thermal output of ^{238}Pu, these isotopes are of primary interest when self-heating is considered. In-growth of 1% ^{241}Am increases the thermal output by about 1 W/kg (Table IV). The thermal outputs of civil plutonium derived from 30 and 60 GW·d/t burnup fuel are approximately 10 and 30 W/kg, respectively.

3.2. PHYSICAL AND CHEMICAL PROPERTIES OF PLUTONIUM

3.2.1. Plutonium metal

Plutonium is a complex metal having six allotropic forms (α, β, γ, δ, δ′, ε) [1]. Some phases exhibit unusual thermal expansion and electrical properties. Plutonium expands upon solidification of the liquid at 640°C. At room temperature, unalloyed (α phase) plutonium has a high density (19.84 g/cm^3), is brittle like cast iron and expands sharply with temperature. When heated from room temperature through the α transition point at 120 ± 2°C, the volume increases by 11%, a change that will rupture containment vessels having insufficient free volume to accommodate the expansion. However, if plutonium is alloyed with elements such as aluminium or gallium, the resulting δ phase metal is malleable and has a more typical expansion coefficient.

Oxidation and other corrosion reactions typically produce small particles containing plutonium and significantly increase the dispersibility hazard posed by the material. Freshly cleaned plutonium is a reactive metal with a lustre similar to nickel [4]. In air, the metal tarnishes and various interference colours appear as the adherent oxide layer becomes thicker. If exposed long enough to oxidizing conditions, an olive green colour appears and loose oxide begins to spall.

The reaction of massive plutonium metal with air at room temperature is slow [5–7]. The oxidation rate depends on a number of factors which include: (a) temperature; (b) surface area; (c) oxygen concentration; (d) the concentrations of moisture and other vapours in the air; (e) the type and extent of alloying; and (f) the presence of a protective layer on the metal surface. The oxidation rate increases with the first four factors and decreases with the last. Alloying can either increase or decrease the oxidation rate, depending on the alloying metal. Oxides formed on surfaces of Pu–Ga alloys are adherent and slow further oxidation. Of all these factors, moisture has a large effect on the oxidation rate and is especially significant in evaluating conditions for storing plutonium metal and oxide. Like iron, the metal is relatively inert in dry air.

Water vapour accelerates the oxidation of plutonium metal by oxygen and reacts directly with the metal [5]. Water inherently reacts with unalloyed metal at a more rapid rate than oxygen at temperatures between –25°C and 200°C and alters the oxidation rate in air at concentrations as low as one part per million [8]. The rate is increased because hydrogen formed by the reaction of water catalytically recombines with oxygen on the oxide surface. The net result is that the oxidation rate is more than a hundred times higher in humid air than in dry air at room temperature. For this reason, plutonium metal has been handled in a dry atmosphere such as one with a –40°C dew point. Nitrogen or argon is effective in reducing the oxidation of plutonium in glove boxes and enclosures if water vapour can also be excluded. Rapid oxidation does not occur if oxygen is present at a level of 5% in nitrogen or argon [9]. However, if 1.3% moisture (50% relative humidity at standard temperature and pressure) accompanies the oxygen, then rapid metal oxidation can be anticipated. Water is not normally used in glove box enclosures containing plutonium metal. The principal source of both oxygen and moisture is diffusion through enclosure gloves and bag-out ports.

Several plutonium oxide compositions form during the oxidation of metal by oxygen or water. Oxide phases corresponding to sesquioxide (Pu_2O_3) and dioxide

(PuO_2) compositions are well characterized [4]. Oxides with stoichiometric compositions between the sesquioxide and dioxide form in liquid water [10]; a superstoichiometric oxide (PuO_{2+x}) is formed by the reaction of water vapour at elevated temperatures [11]. The formation of oxide from metal is accompanied by a 2.5–3.0 fold volume expansion, which may bulge or breach the primary container.

Initiation of a self-sustained reaction of plutonium with air depends on the metal thickness and temperature. Massive metal (greater than 0.2 mm thick) ignites and burns at a constant rate when heated to 500°C [12]. Chips and finely divided powders (less than 0.05 mm thick) burn when heated to 150–250°C because ignition of a pyrophoric Pu_2O_3 layer heats the metal above 500°C. After ignition, plutonium reacts at a rate that sustains continued oxidation [13] and liberates 4.2 kJ of heat per gram of plutonium [14]. The burning temperature depends on the rate of heat dissipation to the surroundings and the rate of heat generation, which is dependent on the surface area of the oxidizing metal. The temperatures of plutonium fires usually exceed the melting point of plutonium (640°C) and may cause unreacted metal to consolidate into a molten configuration.

Plutonium hydride (PuH_x, $2 < x < 3$) forms during the corrosion of plutonium metal by hydrogen from water, organic materials and other sources [15]. The quantity of hydride produced depends on the rate of hydride formation [16] and on the magnitude of the hydrogen containing source. Although the reactivity of plutonium hydride in air depends on factors such as the particle size, absence/presence of a protective oxide layer and the hydrogen to plutonium ratio (x), the hydride is normally pyrophoric in air at room temperature. Thus, the only safe practice is to handle and store hydride or partially hydrided metal in a dry, oxygen free atmosphere and in the absence of unnecessary combustibles, or to convert the material to oxide under controlled conditions.

Plutonium hydride readily reacts with air at room temperature to produce PuO_2 and H_2. PuH_x reacts with nitrogen at 250°C to form plutonium nitride (PuN) and H_2 [17]. When $x = 2$, the reaction with O_2 releases 3.7 kJ of heat per gram of hydride. If sufficient PuH_x is present with plutonium, reaction of the hydride may be sufficient to heat the metal above the 500°C and cause it to spontaneously ignite [12].

Plutonium carbides and nitride, which were considered for use in fast reactors, are highly reactive, especially in the form of fine powders. These materials, which are prepared from the metal, could pose handling problems if exposed to oxygen containing atmospheres, especially air [4]. They react readily with moisture to form gaseous products such as methane, acetylene and ammonia. Since plutonium compounds of this type have been prepared at several sites and may have been ‘temporarily’ stored in sealed vessels without being oxidized, caution should be exercised in opening containers that might contain such materials. Although the reaction of these materials might not be sufficient to ignite plutonium metal, adequate caution is advised.

3.2.2. Plutonium oxide

Plutonium dioxide is derived from two distinctly different sources. The major fraction of oxide from civil sources is prepared by the thermal decomposition of oxalate precipitate or the nitrate. Oxides from weapon sources may in large measure be derived either from oxidation of the metal or from oxidation of hydride prepared from the metal.

The properties of the oxide vary with the method and conditions of preparation [9, 13, 18, 19]. The particle size distribution and specific surface area of the process oxide prepared by pyrolysis or calcination of a precipitate typically differ from those of plutonium dioxide obtained by the oxidation of metal. High temperature (>500°C) oxidation of metal yields an oxide with a relatively low specific surface area (<0.1 m^2/g), a value which is undesirable for fuel fabrication. Products obtained by thermal decomposition of oxalate have high specific areas (10–50 m^2/g) that decrease with increasing calcination temperature [19]. The specific areas of oxides formed by the oxidation of metal in air at room temperature are typically in the 1–20 m^2/g range. The purity of process oxides may also vary with the calcination conditions owing to varying amounts of anion residue remaining in the product after firing [18]. Whereas high surface areas are most suitable for fuel fabrication and low values are desirable for storage, an intermediate surface area of about 10 m^2/g is suitable for both.

Although chemical corrosion is not a concern with PuO_2, reactions involving oxides are known. In addition to the pyrophoric Pu_2O_3 [4], oxides generated by the corrosion of metal in oxygen deficient atmospheres containing hydrogen may include metal fines and small amounts of hydrides. Experience indicates that these products oxidize in a relatively rapid but safe manner. However, this general rule should not be considered applicable if the oxide contains large amounts of potentially reactive impurities such as hydrides. Reactive impurities can pose a fire hazard by acting as an ignition source for combustible materials. Stabilization of these oxides by firing to dioxide composition in air is advisable. The risk of a large fire is virtually eliminated if the amount of combustible material in the storage environment is limited.

Plutonium dioxide is hygroscopic when it has a high surface area to volume ratio, and it adsorbs water up to 1% of its weight under typical laboratory conditions and up to several mass per cent in humid atmospheres [9, 18, 19]. The quantity of water adsorbed is a direct function of the oxide surface area. Adsorbed moisture is removed by heating the oxide in air [19]. A large fraction of the moisture acquired during air exposure is physically adsorbed and can be removed by heating at 50–100°C. Residual moisture levels less than 0.2 mass per cent are obtained by calcining at 700°C. Temperatures up to 1000°C are necessary to completely remove the remaining chemisorbed water. When PuO_2 is heated above 900°C, its specific surface area is always reduced to less than 5 m^2/g, a value that limits readsorption of moisture to about 0.2% of the oxide weight in air at room temperature and 50%

TABLE VI. SOURCE TERMS FOR RESPIRABLE AND DISPERSIBLE FRACTIONS OF OXIDES PREPARED UNDER DIFFERENT CONDITIONS

Conditions of oxide preparation	Cumulative mass fraction	
	3 µm diameter	10 µm diameter
Oxalate decomposition at ≤400°C	1×10^{-5}	1×10^{-2}
Oxidation of Pu in air at 25°C	0.04	0.97
Oxidation of Pu in air at ≥500°C	1×10^{-5}	1×10^{-2}
Oxidation of hydride	6×10^{-3}	0.2

relative humidity. Reduction of the water content is desirable to minimize the potential for pressure generation during extended storage, as well as to diminish neutron moderation and the associated criticality risk.

Plutonium dioxide also adsorbs other atmospheric molecular species that may be reactive from a radiolytic perspective [9]. Together, the adsorption behaviour and the high specific surface areas of process oxides can result in adsorption of large amounts of water, carbon dioxide, organic molecules, etc., from the process environment. The radiolytic decomposition of adsorbed species by alpha particles from the radioactive decay of plutonium generates gases (e.g. H_2 and CO) that are not strongly adsorbed by the oxide. The resulting pressurization of sealed containers can lead to bulging or rupture. Since adsorbed species are removed by heating the oxide in air, thermal processing may be used to remove adsorbates from the oxide prior to storage. Care must also be taken to prevent re-exposure of processed material to moisture and other contaminants before packaging. Thermal desorption of any gas due to internal or external heating of a sealed storage container can generate high pressures. Such pressure generation from stored oxide can be avoided by using the practices described in this report.

The alpha decay of plutonium provides another potential source for pressurizing a sealed container. For example, the decay of ^{239}Pu produces helium at a rate of 0.12 mmol He/kg plutonium per year. Whereas the helium produced in oxide diffuses out of the solid particles, helium generated in bulk metal is retained. As shown by the half-lives for the isotopes in Table IV, He production from ^{238}Pu occurs at a 270 fold faster rate than for ^{239}Pu. The contribution of helium to pressurization of an oxide container is negligible for near term storage, but must be considered during the extended storage of civil plutonium.

Quantitative measures of the potential environmental hazard posed by plutonium oxide are provided by source terms for dispersal and respiration. As determined by the distribution of mass fraction with particle size, the source term data define the mass fractions of PuO_2 that are in the dispersible range, which is less than 10 m geometric particle diameter, and in the respirable range (which is less than about 3 m geometric particle diameter). As shown by the data in Table VI, the respirable and dispersible fractions of oxide vary markedly with process history [13, 19].

3.2.3. Plutonium solutions

The chemistry of plutonium aqueous solutions is very complex [4]. Plutonium has a multiplicity of oxidation states in solution and the equilibrium and kinetic relationship between them results in a very complicated system. For example, plutonium ions in solution can exist in the (III), (IV), (V) and (VI) oxidation states as Pu^{3+}, Pu^{4+}, PuO^{2+} and PuO_2^{2+}, respectively. Because of unique relationships between equilibria and the kinetics of converting from one state to another, it is possible for all four oxidation states to coexist in appreciable concentrations in the same solution. Stabilization of a single ionic species is possible under controlled conditions. Tetravalent plutonium is the most stable state, such as in nitric acid solution. The existence of several oxidation states and the tendency to form complex ions facilitates the separation of plutonium. Pu(VII) also exists as the PuO_3^+ species at high pH values but it is not encountered in processing operations, biological systems or the environment.

Aqueous solutions of plutonium are frequently employed in processing operations. Plutonium nitrate solutions, which are used extensively, are prepared by dissolving metal or oxide in concentrated nitric acid. Dissolution rates are enhanced by adding small amounts of hydrofluoric acid or oxidizing species such as divalent silver. The hydrolysis of tetravalent plutonium in an aqueous solution produces a complex hydroxide polymer [4]. The extent of polymer formation depends on the Pu(IV) concentration, pH, the presence of other ions and temperature. Over time, this bright green polymeric product may settle or precipitate with loss of solution homogeneity and enhancement of the criticality risk. The net hydrolysis process, which is described by the following reaction, has an equilibrium constant, K, of 3.1×10^{-10}:

$$Pu^{4+} + 4H_2O = Pu(OH)_4\ (aq) + 4H^+$$

Solvent extraction is the primary method for separating plutonium from uranium and fission products [4]. The most important extractants for plutonium are organophosphorus chelating compounds such as tri-n-butylphosphate (TBP); ketones such as methylisobutylketone (hexone), ethers and organonitrogen compounds can also be used. As defined by the ratio of the equilibrium plutonium concentrations in the organic and aqueous phases, the coefficient for TBP extraction is large and facilitates production scale application of solvent extraction for processing spent fuels.

Plutonium is easily removed from solution by precipitation as oxalate, hydroxide or peroxide [4]. The removal of Pu(IV) from dilute solutions is accomplished by co-precipitation as hydroxide with Fe(III) or Al(III), but the process is sensitive to the presence of complexing agents and detergents.

3.3. CHEMICAL AND RADIOLYTIC REACTIONS

Chemical changes of materials in processing equipment and storage vessels are promoted by alpha particles and neutron and gamma radiation from the decay of plutonium [3, 9]. The use of organic substances such as plastics, elastomeric O rings, etc., should be avoided if possible because radiolysis by plutonium containing solutions and particles in contact with such materials degrade their properties and generate hydrogen, as well as other non-condensable and reactive gases. Radiolysis in aqueous and organic solutions by dissolved plutonium also produces hydrogen rich gases. Radiolysis of water adsorbed on the oxide may be of concern, even though recent experiments show that the formation of water by catalytic recombination of H_2 and O_2 on the surface of PuO_2 is faster than the radiolytic decomposition of water by ^{239}Pu oxide [8, 20]. In all cases, caution must be exercised to prevent pressurization of equipment and containment failure.

Additional chemical consequences of radiolysis may occur if plutonium metal is present. Hydrides formed by the reaction of plutonium with hydrogen from the radiolytic decomposition of organic materials may ignite spontaneously upon exposure to air [12, 15]. The combination of material degradation, hydride formation and the hydride accelerated oxidation of plutonium is known to cause rapid failure of storage containers for metal and to release plutonium into the environment [21, 22]. The rupture of storage containers and the release of material owing to pressurization by radiolytic gases is well documented for oxides stored in contact with organics [23].

An additional chemical reaction of importance for plutonium handling is the nitration reaction of organic materials by aqueous plutonium nitrate solutions. In the radiolytic environment of these chemical systems, nitration of organic compounds such as ion exchange resins occurs over time. The products obtained after extended periods of time are stable when wet, but are known to violently ignite when dry [4]. Similar concerns exist for nitrate contaminated organic waste, which should be properly packaged and surveyed because of hydrogen generation and a tendency for spontaneous ignition.

The recent characterization of a chemical reaction between water and dioxide [8, 11, 19] suggests that PuO_2 is not thermodynamically stable in air. Water, which accelerates oxidation, reacts with the dioxide as described by the following equation:

$$PuO_2\ (s) + xH_2O\ \text{(adsorbed on oxide)} \rightarrow PuO_{2+x}\ (s) + xH_2\ (g)$$

The binary oxide formed in water vapour at 250°C has a tetragonal structure with a stoichiometry ($x = 0.2$) determined by the fraction of plutonium present as Pu(VI).

The occurrence of this reaction at room temperature is shown by the formation of oxygen free hydrogen by water saturated oxide at a rate of about 1 nmol H_2/m^2 of oxide surface per day [20]. However, additional investigation is necessary to adequately understand this process, to define the potential for significant pressure generation during oxide storage and to determine its relevance to oxide dissolution and formation of high oxidation state plutonium species in aqueous environmental systems.

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4. PLUTONIUM IN THE ENVIRONMENT

4.1. SOURCES OF ENVIRONMENTAL PLUTONIUM

Although estimates indicate that a substantial amount (>4 t) of plutonium has been discharged to the environment, most of that was released prior to 1975. The current rate of plutonium release is very small. The average worldwide activity is low (<70 Bq/m^2) and is contained in the top few centimetres of soil [1]. Plutonium in the environment exists in physical and chemical forms that are immobile and not readily incorporated in biological systems.

Plutonium in the atmosphere has resulted primarily from atmospheric testing of nuclear weapons, the processing of plutonium for such weapons and, to a much lesser extent, from civil reprocessing activities and the destruction of thermoelectric generators on satellites during re-entry to the atmosphere [2]. The amount of plutonium in the environment resulting from weapon fabrication activities during the

TABLE VII. SOURCES AND QUANTITIES OF ATMOSPHERIC PLUTONIUM[a]

Source	Quantity	Alpha emitters				
		^{238}Pu	^{239}Pu	^{240}Pu	^{242}Pu	Total
Weapon testing	Activity (TBq)	330	7400	5200	16	13 000
	Mass (kg)	0.5	3260	590	100	3 950
Satellite accidents	Activity (TBq)	560	—	—	—	560
	Mass (kg)	0.9	—	—	—	0.9
Total civil[b] reprocessing	Activity (TBq)	~300	~700	—	—	1 000
	Mass (kg)	~0.5	~300	—	—	~300
Chernobyl accident	Activity (TBq)	~30	~30	~30	—	~90
	Mass (kg)	<0.1	12	~3	—	~15
Natural sources	Activity (TBq)	—	10	—	—	10
	Mass (kg)	—	~4	—	—	~4
Total	Activity (TBq)	~1200	~8100	~5200	16	~14 600
	Mass (kg)	2	~3600	600	100	~4 300

[a] All data are from Ref. [1] in this section.

[b] The maximum release from civil reprocessing was about 70 TBq per year during the mid-1970s, but it is currently about 0.1 TBq per year.

period is uncertain but is being studied. The estimated quantities and corresponding alpha activities from various sources and isotopes are listed in Table VII.

The quantities of plutonium activity derived from sources other than weapon testing are comparatively small. The total plutonium activity released by the Sellafield facility in the United Kingdom during 40 years of operation is 600–700 TBq, a minor fraction of the total environmental plutonium activity of 14 600 TBq. The estimated contribution from military accidents is also relatively small. For example, the release during the aircraft accident involving nuclear weapons near Thule, Greenland, in 1968 is of the order of 0.9 TBq. Naturally occurring ^{239}Pu formed by the fission of uranium in pitchblende ores is approximately 5 pg/g U and makes a negligible contribution to the total quantity of environmental plutonium.

The deposition of plutonium in the environment has not occurred at a constant rate. The largest increase was experienced during the 1950s and early 1960s from atmospheric testing and weapon fabrication. In the most extreme case, the estimated contribution from a reprocessing facility reached a maximum of about 70 TBq/a during the mid-1970s [2]. Thereafter, the annual release from those facilities decreased steadily to approximately 0.1 TBq/a by the early 1990s. This reduction coincides with a progressive improvement of facilities and procedures. Releases from reprocessing are almost entirely into the sea; releases to the atmosphere are negligible.

Plutonium exists in the environment as oxide, a stable material in most natural media. However, the specific behaviour of plutonium depends on physical and chemical conditions and on available redistribution pathways at the deposition site.

4.2. DISTRIBUTION OF PLUTONIUM IN THE ENVIRONMENT

The distribution of plutonium is not homogeneous over the surface of the earth. Data show that ^{239}Pu concentrations are 70–80 Bq/m^2 at northern latitudes of 35°–45° [2]. The maximum activity in the southern hemisphere is about 15 Bq/m^2 at 25°–45°. The activities of ^{239}Pu are less than 10 Bq/m^2 at the equator and the poles. The higher levels observed in the northern hemisphere correlate with the extent of atmospheric weapon testing conducted at those latitudes. The distribution of ^{238}Pu also varies with latitude, but is essentially equal in the northern and southern hemispheres [2]. A maximum of 3 Bq/m^2 appears near 45°. Activities approach zero at the equator and the poles.

Areas around nuclear test sites, weapon fabrication sites and reprocessing facilities have plutonium activities higher than the values cited above. However, the latitude dependent activities probably reflect amounts of plutonium deposited initially on land and ocean surfaces by fallout.

The plutonium activity at any specific site is difficult to predict because of redistribution processes. Redistribution of land deposited material occurs by the resuspension of particles in the atmosphere, translocation to water and incorporation in the plant–animal food chain. Plutonium deposited in the ocean may dissolve or enter marine life after deposition in sediment on the seabed.

4.3. BEHAVIOUR OF PLUTONIUM IN THE ENVIRONMENT

4.3.1. Behaviour in air

The behaviour of plutonium varies depending on the particle size and dispersal mechanism. Aerosols released in the stratosphere by nuclear weapon tests and satellite re-entry are deposited globally over a period of years. Plutonium released into the atmosphere by fabrication and reprocessing operations and accidents is typically deposited in a relatively short time close to the source. Resuspension of particulate plutonium is not a major concern; fine particles that might again become airborne adhere strongly to the surfaces of large soil particles and aggregates and are not readily entrained. Competition from other translocation processes further reduces the likelihood of resuspension. The likelihood is remote that plutonium containing particles deposited in oceans and other permanent bodies of water will become resuspended (assuming that the bodies retain some water).

4.3.2. Behaviour in soil

Regardless of the source, plutonium in soil is very insoluble and resistant to migration and translocation. The mean migration coefficients for the movement of plutonium in soil are approximately 10^{-7} cm^2/s. The vertical transport rate depends on this coefficient and on the concentration of soluble plutonium species. Since estimates suggest that less than 0.1% of the plutonium in soil is soluble, the concentration gradient driving migration is extremely low, especially since the soluble fraction of plutonium is expected to dissolve over a period of years.

The plutonium concentration is increased by the formation of complex species and by conditions, such as low pH, that hinder the formation of insoluble hydroxide polymers. Local conditions are very important. The presence of wet organic matter tends to increase solubility owing to high humic acid levels. However, clay soils tend to reduce the mobility because their chemically active surfaces adsorb dissolved plutonium containing species.

Translocation of plutonium in soil is primarily lateral. Vertical movement is promoted by certain mechanical processes such as cultivation. Wind and water erosion are the primary mechanisms for the lateral movement of plutonium in soil. Fine silt clay particles contain the highest concentrations of plutonium and are readily transported by water, or by wind when dry.

Biological systems provide additional pathways for the translocation of plutonium from soil. The plutonium fraction transferred to plants after deposition on the leaves is approximately 10^{-5}. For expected concentrations of plutonium in soil, the fraction taken up by roots ranges from 10^{-3} to 10^{-5}. Root uptake of soluble plutonium species in the soil is thought to involve complexes of Pu(VI).

Ingestion and inhalation are the principal mechanisms for the translocation of plutonium from soil to animals, but neither constitutes a significant risk. Inhalation is of minimal concern because airborne concentrations of plutonium are low except in the vicinity of an accident and then only for a short time following the accident. Estimates for a severe occurrence involving high altitude dispersal of oxide suggest that the resulting maximum concentration of respirable particles in the air is of the order of 10^{-7} g/m^3 [3]. Gastrointestinal and pelt uptake by grazing animals over a long period of time is also of minimal concern because the amount of plutonium ingested by animals is small and the fraction of ingested plutonium absorbed by animals is approximately 10^{-4}. Combination of the selectivities of plant and animal systems shows that the fraction of deposited plutonium translocated to herbivores is in the 10^{-7} to 10^{-9} range.

4.3.3. Behaviour in water

Plutonium deposited in fresh water establishes a distributional relationship between the concentration of plutonium in water and the concentration in sediments or particulate matter [4]. Distribution constants, which are in the 10^{-4} to 2×10^{-6} range, show that the plutonium resides primarily as an insoluble solid. The process seems to involve an equilibrium between Pu(III) or Pu(IV) in the solid and Pu(V) or Pu(VI) in solution. The highest concentrations in solution are observed in systems with low pH and high sulphate concentration. As noted in a recent assessment of the hazard posed by plutonium in the environment [3], plutonium oxide is less soluble in water than ordinary sand (SiO_2) and becomes immobile after settling and entrapment in sediments.

The distribution behaviour of plutonium in marine systems is very similar to that in fresh water. Coefficients for equilibration of sedimentary and dissolved plutonium are also between 10^{-4} and 10^{-5}, suggesting that the chemical processes in salt water are similar to those in fresh water. Translocation of plutonium in lakes and oceans occurs by chemical and mechanical processes. Both vertical and lateral transport processes are possible, depending on the conditions. Studies of plutonium uptake by marine organisms show that concentration factors relative to the plutonium concentration in sea water are 40, 300 and 3000 for edible portions of fish, crustacea and mollusks, respectively.

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5. PATHWAYS TO HUMANS AND THE BIOLOGICAL EFFECTS OF PLUTONIUM

5.1. INTERNAL EXPOSURE TO PLUTONIUM

After more than 50 years of investigation, the biochemistry and toxicology of plutonium are far better known than for most other elements and radionuclides, and the metabolism of plutonium and related elements has been extensively reviewed in a number of documents [1–6].

Plutonium is handled as an extremely hazardous and toxic radioactive material; however, its chemical toxicity is inconsequential when compared with its radiotoxicity. The biological effects of plutonium inside the human body are due primarily to the alpha radiation emitted by the isotopes of plutonium and by the gamma radiation from ^{241}Am, which is a daughter product of ^{241}Pu. The magnitude of the alpha radiation dose depends on the activity of plutonium involved, on the physical and chemical form of the plutonium, on the specific activity of the plutonium and on the route of intake. Plutonium contamination on the outside of the human body delivers a negligible radiation dose because the alpha radiation does not have enough energy to penetrate the skin [7].

From the point of view of industrial radiation protection, oxides are probably the most important class of compounds. The oxides found in industrial situations may be in pure form or in non-stoichiometric mixtures with other oxides; they may have been formed at either very high temperatures (above 1500°C) or relatively low temperatures (including ambient). The composition and formation temperature of the oxides can markedly influence their solubility and, thus, their biological behaviour. In MOX fuel, the plutonium oxide may be combined with a greater mass of uranium oxide, which is more soluble than the plutonium oxide. In vivo, this bulk matrix is relatively rapidly solubilized, leaving the plutonium oxide in the form of very small aggregates, <1 nm in diameter, which may not dissolve completely, but appear to enter the systemic circulation and to be deposited in tissues or excreted in particulate form [1].

Plutonium intake in humans may occur via inhalation, through wounds, or via ingestion (oral intake). Inhalation is undoubtedly the most significant route of occupational exposure [1]. Inhalation of significant quantities of plutonium can occur in a matter of moments, usually due to a breach of containment or loss of ventilation control resulting in transient but high airborne concentrations of plutonium. As an example, inhalation of 1 g of plutonium oxide from a military source 15 years after reprocessing (with an estimated specific activity of 3 × 10^9 alpha Bq/g (0.08 alpha Ci/g)) results in a committed effective dose of about 0.04 Sv (using the dose conversion factors of Table VIII) [8].

Uptakes via wounds can also be of major concern because dosimetrically significant quantities of plutonium can be absorbed from the wound directly into the bloodstream. All wounds contaminated with plutonium should be treated as serious and may warrant treatment with chelating agents or by excision.

Ingestion is a fairly minimal concern. Human gut transfer factors for plutonium compounds are given in Tables VIII and IX [8]. It is unlikely that a person will ingest large quantities of plutonium and, if ingested, the material is not efficiently absorbed through the gastrointestinal (GI) tract. For example, if 1 g of plutonium oxide with a specific activity of 3 × 10^9 alpha Bq/g is ingested, the committed effective dose is approximately 0.03 mSv. In comparison, natural background radiation results in annual doses of the order of 2–4 mSv. When dissolved in certain solvents, plutonium can be absorbed through the skin. However, this route of intake is uncommon.

The estimated absorbed doses from civil and military plutonium are similar when measured on an activity basis, but when measured on a mass basis, the dose for civil plutonium is a factor of about ten greater than for military material because of the higher specific activity, due primarily to ^{238}Pu. The specific alpha activity of plutonium with a known initial isotopic composition and age is readily calculated using data in Table IV and Fig. 3. Accordingly, the committed effective dose for civil plutonium is also about ten times higher than for military plutonium.

Intakes of quantities of plutonium may produce delayed health effects, such as increased risk of cancer mortality, and delayed or acute (stochastic) health effects, such as pulmonary oedema. However, relatively high doses are required to produce acute effects. For example, about 20 mg of optimally sized (roughly 1 µm) plutonium would have to be inhaled to cause death within roughly a month from pulmonary fibrosis or pulmonary oedema, and about 500 mg of plutonium with a specific activity of 3 × 10^9 alpha Bq/g would have to be ingested to deliver a lethal dose [7]. In practice, ingested plutonium is less acutely toxic than many common poisons such as strychnine, lead arsenate and cyanide. Even with inhalation, the acute toxicity of plutonium is similar to that of heavy metal vapours like cadmium and mercury. The levels at which acute effects might occur are orders of magnitude above the levels encountered in normal occupational situations, where typical body activities range from non-detectable to less than 100 Bq [9]. There are no known cases of human mortality from plutonium inhalation or ingestion.

Alpha irradiation of lungs and other organs is assumed to increase the likelihood of cancer at the sites of high plutonium concentration. The statistical (stochastic) effect of an increased cancer risk factor (when expressed across a large exposed population) is estimated to be one additional fatal cancer induced for each

TABLE VIII. WORKER’S COMMITTED EFFECTIVE DOSE PER UNIT INTAKE VIA INHALATION AND INGESTION {**e(g)**[a]} (Sv·Bq^{-1}) FROM VARIOUS PLUTONIUM ISOTOPES AND ^{241}Am [7]

Nuclide	Half-life (a)	Inhalation				Ingestion	
		Type[b]	f_1[c]	$\mathbf{e(g)}_{1\ \mu m}$[a, d]	$\mathbf{e(g)}_{5\ \mu m}$[a, d]	f_1[c]	**e(g)**[a]
^{236}Pu	2.85	M	5.0×10^{-4}	$1.8 \times 10.^{-5}$	1.3×10^{-5}	5.0×10^{-4}	8.6×10^{-8}
		S	1.0×10^{-5}	9.6×10^{-6}	7.4×10^{-6}	1.0×10^{-5}	6.3×10^{-9}
						1.0×10^{-4}	2.1×10^{-8}
^{238}Pu	87.7	M	5.0×10^{-4}	4.3×10^{-5}	3.0×10^{-5}	5.0×10^{-4}	2.3×10^{-7}
		S	1.0×10^{-5}	1.5×10^{-5}	1.1×10^{-5}	1.0×10^{-5}	8.8×10^{-9}
						1.0×10^{-4}	4.9×10^{-8}
^{239}Pu	2.41×10^{4}	M	5.0×10^{-4}	4.7×10^{-5}	3.2×10^{-5}	5.0×10^{-4}	2.5×10^{-7}
		S	1.0×10^{-5}	1.5×10^{-5}	8.3×10^{-6}	1.0×10^{-5}	9.0×10^{-9}
						1.0×10^{-4}	5.3×10^{-8}
^{240}Pu	6.54×10^{3}	M	5.0×10^{-4}	4.7×10^{-5}	3.2×10^{-5}	5.0×10^{-4}	2.5×10^{-7}
		S	1.0×10^{-5}	1.5×10^{-5}	8.3×10^{-6}	1.0×10^{-5}	9.0×10^{-9}
						1.0×10^{-4}	5.3×10^{-8}
^{241}Pu	14.4	M	5.0×10^{-4}	8.5×10^{-7}	5.8×10^{-7}	5.0×10^{-4}	4.7×10^{-9}
		S	1.0×10^{-5}	1.6×10^{-7}	8.4×10^{-8}	1.0×10^{-5}	1.1×10^{-10}
						1.0×10^{-4}	9.6×10^{-10}
^{242}Pu	3.76×10^{5}	M	5.0×10^{-4}	4.4×10^{-5}	3.1×10^{-5}	5.0×10^{-4}	2.4×10^{-7}
		S	1.0×10^{-5}	1.4×10^{-5}	7.7×10^{-6}	1.0×10^{-5}	8.6×10^{-9}
						1.0×10^{-4}	5.0×10^{-8}
^{241}Am	432	M	5.0×10^{-4}	3.9×10^{-5}	2.7×10^{-5}	5.0×10^{-4}	2.0×10^{-7}

TABLE VIII. (cont.)

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[a] The committed effective dose per unit intake {**e(g)**} is the sum of the doses to all organs and tissues, weighted by their sensitivities to radiation and integrated over a lifetime, from the intake of unit activity of a radionuclide.

[b] For inhalation of particulates, types M and S denote, respectively, moderate and slow clearance from the lung. For plutonium, insoluble oxides are S; all other compounds are M; for americium, all compounds are M.

[c] The gut transfer factor f_1 represents the portion of the intake transferred to body fluids in the gut. For inhalation the f_1 values are valid for the component of the intake cleared from the lung to the GI tract. For compounds of plutonium, the gut transfer factor f_1 is: 1×10^{-4} for nitrates, 1×10^{-5} for insoluble oxides, and is taken as 5.0×10^{-4} for all other compounds. For compounds of americium, the gut transfer factor f_1 is 5×10^{-4} for all compounds.

[d] The 1 and 5 μm subscripts of the committed effective dose per unit intake {e(g)} ‘represent’ the diameters of the particles.

TABLE IX. COMMITTED EFFECTIVE DOSE PER UNIT INTAKE {**e(g)**}[a] VIA INGESTION (Sv·Bq^{-1}) FOR MEMBERS OF THE PUBLIC [7]

Nuclide	Half-life (a)	Age ≤ 1 a		f_1[b] for age >1 a	Age				
		f_1[b]	**e(g)**[a]		1–2 a **e(g)**[a]	2–7 a **e(g)**[a]	7–12 a **e(g)**[a]	12–17 a **e(g)**[a]	>17 a **e(g)**[a]
^{236}Pu	2.85	0.005	2.1×10^{-6}	5.0×10^{-4}	2.2×10^{-7}	1.4×10^{-7}	1.4×10^{-7}	8.5×10^{-8}	8.7×10^{-8}
^{238}Pu	87.7	0.005	4.0×10^{-6}	5.0×10^{-4}	4.0×10^{-7}	3.1×10^{-7}	2.4×10^{-7}	2.2×10^{-7}	2.3×10^{-7}
^{239}Pu	2.41×10^{4}	0.005	4.2×10^{-6}	5.0×10^{-4}	4.2×10^{-7}	3.3×10^{-7}	2.7×10^{-7}	2.4×10^{-7}	2.5×10^{-7}
^{240}Pu	6.54×10^{3}	0.005	4.2×10^{-6}	5.0×10^{-4}	4.2×10^{-7}	3.3×10^{-7}	2.7×10^{-7}	2.4×10^{-7}	2.5×10^{-7}
^{241}Pu	14.4	0.005	5.6×10^{-8}	5.0×10^{-4}	5.7×10^{-9}	5.5×10^{-9}	5.1×10^{-9}	4.8×10^{-9}	4.8×10^{-9}
^{242}Pu	3.76×10^{5}	0.005	4.0×10^{-6}	5.0×10^{-4}	4.0×10^{-7}	3.2×10^{-7}	2.6×10^{-7}	2.3×10^{-7}	2.4×10^{-7}
^{241}Am	432	0.005	3.7×10^{-6}	5.0×10^{-4}	3.7×10^{-7}	2.7×10^{-7}	2.2×10^{-7}	2.0×10^{-7}	2.0×10^{-7}

[a] The committed effective dose per unit intake {**e(g)**} is the sum of the doses to all organs and tissues, weighted by their sensitivities to radiation and integrated over a lifetime, from the intake of unit activity of a radionuclide.

[b] The gut transfer factor f_1 represents the portion of the intake transferred to body fluids in the gut.

0.14 mg of plutonium inhaled[1] and one additional fatal cancer induced for each 30 mg of plutonium ingested[2], assuming a specific activity of 3 × 10^9 alpha Bq/g.

5.2. ENTRY OF PLUTONIUM BY INHALATION

The ‘Human Respiratory Tract Model for Radiological Protection’ [10] updates the model used in ICRP Publication 30 [11]. The new model takes into account extensive data on the behaviour of inhaled materials and, as in the earlier model, deposition and clearance are treated separately. Whereas the earlier model calculates only the average dose to the lungs, the new model calculates doses to specific tissues of the respiratory tract, and thus takes account of differences in radiosensitivity within the tract.

Section 2 of ICRP Publication 68 [13] provides a good summary of the new respiratory tract model, the details of which are found in ICRP Publication 66 [10]. From an operational standpoint, two major factors are of concern:

- The initial pattern of deposition within the different regions of the respiratory tract (and subsequent dose distribution) is dependent upon the particle size distribution of the airborne activity.
- The subsequent retention and distribution of deposited activity is determined by the chemical form of the plutonium. Oxides are retained in the lungs for years, whereas more transportable compounds are transferred much more quickly into systemic distribution.

Following deposition in the lung, material is cleared at a rate dependent upon its location, chemical form and mode of deposition. Material deposited directly in a region during inhalation is cleared at a different rate than that which is cleared into a region following deposition in another region. Material cleared from the lung is transported to the GI tract, the lymph nodes and the blood; absorption into blood depends on the physical and chemical form of the deposited material. The

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[1] *For inhalation:* (0.05 fatal cancers/Sv (ICRP 60) [12]) × (4.7 × 10^{-5} Sv/Bq) (Table VIII) × (3 × 10^9 Bq/g) (Table IV) = 7 × 10^3 fatal cancers/g. Or ~0.14 mg of Pu inhaled per fatal cancer induced. The largest speck of plutonium that can be readily inhaled is about 3 μm in diameter and has a mass of about 0.14 millionths of a milligram (1.4 μm × 10^{-10} g). The risk of developing a fatal cancer as a result of inhaling such a particle of plutonium is ~10^{-6}.

[2] *For ingestion:* (0.05 fatal cancers/Sv (ICRP 60) [12]) × (2.5 × 10^{-7} Sv/Bq) (Table VIII) × (3 × 10^9 Bq/g) (Table IV) = 37.5 fatal cancers/g. Or ~30 mg of Pu ingested per fatal cancer induced.

rate of clearance from the lung will affect the total dose delivered and material specific rates of absorption should be used when reliable human or animal data exist.

In the lung model given in Ref. [10], compounds are classified according to their clearance from the lung as fast (type F), moderate (M), or slow (S). These clearance rates correspond roughly to the ICRP Publication 30 clearance rates of daily (D), weekly (W), or yearly (Y) (Ref. [11], pp 5–9.) Plutonium oxides are typically in clearance class S. All other compounds of plutonium may be tentatively assumed to act as type M materials (Ref. [11], Table F.1).

Plutonium may have different clearance characteristics when mixed with oxides of other metals, such as Pu–Ga alloys, or when in the presence of sodium or potassium oxides (see Ref. [14], Section 5.29, for a further discussion on absorption types). The proposed translocation rate for plutonium oxide is between 0.02% and 0.1% per day. Studies of the most important soluble plutonium compounds, namely the nitrate and TBP complex, indicate a more rapid plutonium translocation from the lungs. The rates may vary considerably from the value of 0.5% per day used in models.

Table VIII, extracted from IAEA Safety Series No. 115 [8], provides the committed effective dose per unit intake via inhalation and ingestion for workers from various plutonium isotopes. Tables IX [7] and X [7, 14] provide similar data for members of the public for intake by ingestion and inhalation, respectively.

5.3. GASTROINTESTINAL ABSORPTION OF PLUTONIUM

Plutonium may enter the GI tract following ingestion or inhalation. Up to 50% of inhaled insoluble compounds may be cleared from the lung via the GI tract. When plutonium enters the GI tract, a fraction of the material passes through the cells of the mucosa into the bloodstream. A very small fraction of the ingested material is transferred to the blood and is called the ‘fractional absorption’ (f_1) [8]. Immediately after entry into the bloodstream, a part of the absorbed material is excreted in the urine and faeces (via the bile) (Ref. [15], p. 21). Absorption can be further reduced by appropriate countermeasures (e.g. chelation). The quoted fractional transfers for adults lie in the range of 10^3 to 10^5 depending on the nature of the ingested material.

The absorption of plutonium compounds in various animal species depends on many factors, including experimental conditions. In all situations where the characteristics of the plutonium intake cannot be described precisely, an absorption fraction of 10^3 gives a sufficient margin of safety for radiation protection purposes. Smaller values should be used for occupational exposure, where the chemical and physical state of the ingested material can be confidently established. The current ICRP Publication 68 recommendation of 10^4 and 10^5 remains appropriate for plutonium nitrate and insoluble oxide, respectively [13]. For compounds with some valence states of plutonium, GI absorption is altered when large amounts (>1 mg/kg) are

TABLE X. COMMITTED EFFECTIVE DOSE PER UNIT INTAKE {**e(g)**[a]} VIA INHALATION (Sv·Bq^{-1}) FOR MEMBERS OF THE PUBLIC [7]

Nuclide	Half-life (a)	Type[b]	Age ≤ 1 a		f_b^1 for age >1 a			Age		
			f_1[c]	**e(g)**[a]		1–2 a **e(g)**[a]	2–7 a **e(g)**[a]	7–12 a **e(g)**[a]	12–17 a **e(g)**[a]	>17 a **e(g)**[a]
^{236}Pu	2.85	F	0.005	1.0×10^{-4}	5.0×10^{-4}	9.5×10^{-5}	6.1×10^{-5}	4.4×10^{-5}	3.7×10^{-5}	4.0×10^{-5}
		M	0.005	4.8×10^{-5}	5.0×10^{-4}	4.3×10^{-5}	2.9×10^{-5}	2.1×10^{-5}	1.9×10^{-5}	2.0×10^{-5}
		S	1.0×10^{-4}	3.6×10^{-5}	1.0×10^{-5}	3.1×10^{-5}	2.0×10^{-5}	1.4×10^{-5}	1.2×10^{-5}	1.0×10^{-5}
^{238}Pu	87.7	F	0.005	2.0×10^{-4}	5.0×10^{-4}	1.9×10^{-4}	1.4×10^{-4}	1.1×10^{-4}	1.0×10^{-4}	1.1×10^{-4}
		M	0.005	7.8×10^{-5}	5.0×10^{-4}	7.4×10^{-4}	5.6×10^{-5}	4.4×10^{-5}	4.3×10^{-5}	4.6×10^{-5}
		S	1.0×10^{-4}	4.5×10^{-5}	1.0×10^{-5}	4.0×10^{-5}	2.7×10^{-5}	1.9×10^{-5}	1.7×10^{-5}	1.6×10^{-5}
^{239}Pu	2.41×10^{4}	F	0.005	2.1×10^{-4}	5.0×10^{-4}	2.0×10^{-4}	1.5×10^{-4}	1.2×10^{-4}	1.1×10^{-4}	1.2×10^{-4}
		M	0.005	8.0×10^{-5}	5.0×10^{-4}	7.7×10^{-5}	6.0×10^{-5}	4.8×10^{-5}	4.7×10^{-5}	5.0×10^{-5}
		S	1.0×10^{-4}	4.3×10^{-5}	1.0×10^{-5}	3.9×10^{-5}	2.7×10^{-7}	1.9×10^{-5}	1.7×10^{-5}	1.6×10^{-5}
^{240}Pu	6.54×10^{3}	F	0.005	2.1×10^{-4}	5.0×10^{-4}	2.0×10^{-4}	1.5×10^{-4}	1.2×10^{-4}	1.1×10^{-4}	1.2×10^{-4}
		M	0.005	8.0×10^{-5}	5.0×10^{-4}	7.7×10^{-5}	6.0×10^{-5}	4.8×10^{-5}	4.7×10^{-5}	5.0×10^{-5}
		S	1.0×10^{-4}	4.3×10^{-5}	1.0×10^{-5}	3.9×10^{-5}	2.7×10^{-5}	1.9×10^{-5}	1.7×10^{-5}	1.6×10^{-5}
^{241}Pu	14.4	F	0.005	2.8×10^{-6}	5.0×10^{-4}	2.9×10^{-6}	2.6×10^{-6}	2.4×10^{-6}	2.2×10^{-6}	2.3×10^{-6}
		M	0.005	9.1×10^{-7}	5.0×10^{-4}	9.7×10^{-7}	9.2×10^{-7}	8.3×10^{-7}	8.6×10^{-7}	9.0×10^{-7}
		S	1.0 × 10–4	2.2 × 10–7	1.0 × 10–5	2.3 × 10–7	2.0 × 10–7	1.7 × 10–7	1.7 × 10–7	1.7 × 10–7

Continued on next page.

TABLE X. COMMITTED EFFECTIVE DOSE PER UNIT INTAKE {**e(g)**[a]} VIA INHALATION ($Sv \cdot Bq^{-1}$) FOR MEMBERS OF THE PUBLIC [7] (cont.)

Nuclide	Half-life (a)	Type[b]	Age ≤ 1 a		f_b^1 for age >1 a	Age				
			f_1[c]	**e(g)**[a]		1–2 a **e(g)**[a]	2–7 a **e(g)**[a]	7–12 a **e(g)**[a]	12–17 a **e(g)**[a]	>17 a **e(g)**[a]
^{242}Pu	3.76×10^{5}	F	0.005	2.0×10^{-4}	5.0×10^{-4}	1.9×10^{-4}	1.4×10^{-4}	1.2×10^{-4}	1.1×10^{-4}	1.1×10^{-4}
		M	0.005	7.6×10^{-5}	5.0×10^{-4}	7.3×10^{-5}	5.7×10^{-5}	4.5×10^{-5}	4.5×10^{-5}	4.8×10^{-5}
		S	1.0×10^{-4}	4.0×10^{-5}	1.0×10^{-5}	3.6×10^{-5}	2.5×10^{-5}	1.7×10^{-5}	1.6×10^{-5}	1.5×10^{-5}
^{241}Am	432	F	0.005	1.8×10^{-4}	5.0×10^{-4}	1.8×10^{-4}	1.2×10^{-4}	1.0×10^{-4}	9.2×10^{-5}	9.6×10^{-5}
		M	0.005	7.3×10^{-5}	5.0×10^{-4}	6.9×10^{-5}	5.1×10^{-5}	4.0×10^{-5}	4.0×10^{-5}	4.2×10^{-5}
		S	0.005	4.6×10^{-5}	5.0×10^{-4}	4.0×10^{-5}	2.7×10^{-5}	1.9×10^{-5}	1.7×10^{-5}	1.6×10^{-5}

[a] The committed effective dose per unit intake {**e(g)**} is the sum of the doses to all organs and tissues, weighted by their sensitivities to radiation and integrated over a lifetime, from the intake of unit activity of a radionuclide.

[b] For inhalation of particulates, types F, M and S denote, respectively, fast, moderate and slow clearance from the lung. For plutonium, insoluble oxides are S; all other compounds are M; for americium, all compounds are M. When no specific information is available, the recommended default absorption type for particulate aerosols is M (see Ref. [14]).

[c] The gut transfer factor f_1 represents the portion of the intake transferred to body fluids in the gut. For inhalation, the f_1 values are valid for the component of the intake cleared from the lung to the GI tract. For compounds of plutonium, the gut transfer factor f_1 is: 1×10^{-4} for nitrates, 1×10^{-5} for insoluble oxides and is taken as 5.0×10^{-4} for all other compounds. For compounds of americium, the gut transfer factor f_1 is 5×10^{-4} for all compounds.

administered [16]. Absorption of ^{241}Am lies between 4×10^5 and 6×10^4 for all types of ingested compounds.

5.4. PENETRATION OF PLUTONIUM THROUGH INTACT SKIN

Skin contamination may occur during abnormal handling operations from accidental releases of plutonium particles or solutions. In most cases, plutonium oxide is easily removed from the skin without negative health effects. Typical methods of decontamination include washing with soap and water, using a soft brush, or removing dead skin. In all cases, decontamination should be discontinued prior to causing any degradation of the skin as the skin provides an effective barrier against the uptake of plutonium. Evidence suggests that putting a rubber glove on a contaminated hand for an extended time and allowing a person to ‘sweat the contamination out’ results in increased absorption, and therefore should not be utilized as a decontamination technique.

Limited human experience suggests that the absorption of plutonium from a dilute aqueous acid solution probably does not exceed 0.01% during the first hour after contamination. The absorption of plutonium from the TBP complex may be 0.04% within 15 min. Increased radionuclide absorption into the body may result from skin damage caused by strong acids or from complexing agents used in skin decontamination.

After penetrating the skin, the largest amount of plutonium in a nitrate solution is found in the bones and the liver. Thus, absorbed plutonium displays the typical distribution found for other soluble compounds entering the body by other routes.

5.5. DISTRIBUTION AND RETENTION OF ABSORBED PLUTONIUM

Once plutonium enters the bloodstream, approximately 80% of the element deposits in the skeleton and liver. The remaining 20% is excreted, or deposited in a variety of other organs and tissues where the concentrations of plutonium do not approach those in the liver or bone. A possible exception is the gonads, which require separate consideration for evaluation of genetic risk. The complicated biological mechanism of plutonium retention, distribution and clearance is described by the formal mathematical models for radiation protection. These metabolic models are presented in ICRP Publications 67 [17] and 68 [13].

The partitioning of plutonium between liver and bone varies widely from individual to individual. After absorption in the body, plutonium shows an average deposition of 50% in the skeleton and 30% in the liver. However, the demonstrated individual variability is such that continued use of the assumptions employed in ICRP

Publication 30, of 90% total deposition with 45% in the liver and 45% in the skeleton, remains an appropriately cautious procedure for radiation protection purposes (Ref. [11], p. 75.)

A considerable body of data from animal studies and qualitative observations for humans indicates that irradiation of the foetus is not a critical factor in controlling the occupational exposure of pregnant women (Ref. [1], para. 6.77). Actinides do not concentrate preferentially in the foetus and such deposition is rapidly diluted by growth. Control procedures that effectively protect the mother will amply protect the foetus, even though the foetus may have a greater sensitivity to radiation.

5.6. REDUCING THE DOSE FROM INTERNALLY DEPOSITED PLUTONIUM

In serious or extreme circumstances, medical intervention for intakes of plutonium may be warranted. Such procedures may include administration of a chelating agent, wound excision or, in very extreme cases, lung lavage. The decision criteria established to direct actions in such cases must balance the risk of intervention against the potential benefit. Such medical procedures should be performed on an 'informed consent' basis, and the decision to intervene should be jointly made with input from the patient, medical staff and health physics personnel [18].

5.6.1. Chelation therapy

The timely administration of chelating agents can significantly reduce the radiation dose received from an intake of plutonium. Chelating agents are chemicals which combine with metal ions to form stable and soluble molecules. Reaction with transuranic elements such as plutonium in the bloodstream enhances the body's ability to excrete these elements via the urine. The chelating agent currently recommended for use with plutonium is diethylene triamine penta acetic acid, or DTPA, in the form of a calcium (Ca–DTPA) or zinc (Zn–DTPA) compound.

DTPA can dramatically increase the rate of excretion of plutonium from the body, thereby significantly lowering the total dose received. The effectiveness of DTPA treatment depends greatly upon the route of intake of the plutonium, its chemical and physical form, and the time and duration of the DTPA treatment(s). Dose reductions from 10 to 90% have been achieved for wound and burn cases and dose reductions of up to 30% have been achieved for inhalation intakes. DTPA usage at Sellafied, in the United Kingdom, was reviewed in 1994 [19].

To be most effective, the initial administration of DTPA should be given as soon as possible after the suspected intake and preferably within one hour. The normal route of administration of DTPA is via intravenous injection or inhalation of an aerosol. Early administration allows the DTPA to work while the largest quantity of plutonium is still in the bloodstream. DTPA has been used for approximately 30 years and has been successful on hundreds of people. However, since it has little commercial use outside the plutonium handling community, DTPA is still categorized as an ‘Investigational New Drug’ by the US Food and Drug Administration (FDA). As such, its use in the USA is carefully monitored and controlled.

Administration of DTPA is not expected to cause any serious side effects or risks. Some individuals have reported experiencing minor temporary side effects, such as nausea, vomiting, diarrhoea, chills, fever, itching and muscle cramps after multiple administrations. No serious long term effects of DTPA have been observed in humans, other than the depletion of zinc, which can be avoided by using Zn–DTPA for longer term therapy. Animal studies (using doses of Ca–DTPA 50 times higher than the normal doses) show damage to the kidneys and the liver.

Calcium–DTPA may be harmful to a developing foetus. Studies with pregnant mice, which received five daily injections of 20–80 times the normal dose of Ca–DTPA during gestation, show severe foetal injury and death. However, since the studies that use high dosages have only been carried out on animals, the effects on the human foetus of Ca–DTPA are uncertain. Even so, it would be prudent to use Zn–DTPA instead.

5.6.2. Wound excision

With a contaminated wound, a significant amount of plutonium may be present locally at the wound site. Surgical removal (excision) of contaminated tissue can be a very effective method of reducing the dose from such wounds. Decisions about performing such excisions should be based on the physical circumstances of the incident, the results of wound counting measurement (if available), the projected doses and the location, extent and nature of the tissue to be excised.

Recommendations found in the literature regarding excision action levels are summarized below:

- “Wounds containing over 150 Bq (4 nCi) of plutonium should be serious candidates for excision.” [20].
- For wounds containing between 150 Bq (4 nCi) and 75 Bq (2 nCi) of plutonium, the decision to excise should depend on the age of the victim, the location of the wound, the likelihood of successful removal of significant quantities of plutonium, etc.
- It is considered that no significant risk would be incurred if the amount (of plutonium left in the wound site) was less than 75 Bq (2 nCi) [21].

Dose reduction factors of 100 or greater have been achieved by excising contaminated tissue from wounds. Note that if wound excision and chelation therapy are both to be performed, chelation therapy should be administered *before* any attempt at excision because that process can result in a further transfer of activity from the wound site to the bloodstream. ‘Loading’ the bloodstream with DTPA before excision minimizes the ultimate radiation dose.

The risk of tissue excision depends greatly upon the location of the wound, and the nature and quantity of tissue involved. Most excisions involve relatively small amounts of tissue. In all cases, the risk of loss of function or disfigurement must be carefully weighed against the potential reduction in radiation dose. Facilities in which such wounds are possible should have pre-established action guidelines and access to appropriate wound counting equipment.

5.6.3. Lung lavage

Inhalation of a large amount ($>10^5$ Bq (2.7 μCi)) of insoluble plutonium activity, such as might occur in a fire or explosion, can result in serious acute health effects. Intakes on the order of 100 ALI (annual limit of intake) are expected to significantly increase the risk of cancer mortality, and intakes larger than this may result in immediate death. Lung lavage, washing the lungs and airways with isotonic fluids while the patient is under general anaesthesia, is effective in removing up to 50% (more with multiple treatments) of inhaled insoluble materials. The risk associated with this procedure is mainly that of general anaesthesia, which is estimated to be about 0.2% (0.002) per lavage [21].

Recommendations in the CEC/DOE Guidebook state that lung lavage is “justified” or should be “considered” if the intake is estimated to be greater than 100 times the ALI [18]. This intake corresponds approximately to the level at which the increased risk of cancer is significant.

5.7. EXTERNAL EXPOSURE TO PLUTONIUM

The external dose from plutonium arises from both gamma and neutron radiation. The isotopic composition of the plutonium determines the surface dose rates. ^{241}Am contributes most significantly to the gamma dose, and ^{238}Pu and ^{240}Pu contribute most significantly to the neutron dose. The dose from accumulated material (e.g. in HEPA filters) can be a significant source of external exposure and should be monitored on a periodic basis.

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6. LICENSING, CONTROLS AND REGULATORY LIMITS

6.1. LICENSING

Operations involving civil plutonium are permitted only in facilities that are licensed for that purpose. Licensing generally applies throughout the service life of an installation, starting from siting through construction, commissioning, operation, modification and on to eventual decommissioning.

National regulatory agencies achieve the control of nuclear and radiological safety for licensed plants by:

- Carrying out safety assessments of the proposed site and the nuclear plant design;
- Ensuring that the licensees establish safety requirements based on national standards for the protection of workers, members of the public and the environment;
- Conducting inspections for compliance with these requirements at all stages, from construction to operation and eventual decommissioning.

6.2. EXEMPTION LEVELS

The exempt activity concentration and activity levels of the various plutonium isotopes are given in Table XI [1]. Users may be exempted from licensing and reporting requirements by regulatory authorities for sources below these levels.

6.3. OCCUPATIONAL DOSE LIMITS

The ICRP dose limitation system as put forth in ICRP Publication 26 [2] and the Basic Safety Standards (BSS), published in 1982 as IAEA Safety Series No. 9 [3], were adopted by most countries in the 1980s and the dose limits were incorporated in their regulatory documents on radiation protection of the workers and the public. Safety Series No. 9 is now obsolete and has been replaced by the ‘International Basic Safety Standards for Protection Against Ionizing Radiation and for the Safety of Radiation Sources’ (BSS) [1]. Some countries are still using the recommendations contained in Safety Series No. 9 [3], but most are now reviewing their standards in light of ICRP Publication 60 [4] and the new BSS [1]. The latter incorporates the recommendations of ICRP Publication 60. It is envisaged that the revised dose limits will soon be adopted in a phased manner in all countries. Backfitting/modification of the operating plants may be required in order to comply with the new dose limits, as recommended by the ICRP and the IAEA. As a result of intensive radiation

TABLE XI. EXEMPT ACTIVITY CONCENTRATIONS AND EXEMPT ACTIVITIES OF RADIONUCLIDES (ROUNDED)[a] [1]

Nuclide	Activity concentration (Bq/g)	Activity (Bq)
^{238}Pu	1	1×10^4
^{239}Pu	1	1×10^4
^{240}Pu	1	1×10^3
^{241}Pu	100	1×10^5
^{242}Pu	1	1×10^4

[a] The guidance exemption levels are subject to the following considerations. (a) They have been derived using a conservative model based on: (i) the effective dose expected to be incurred by any member of the public due to the exempted practice or source is of the order of 10 μSv or less in a year, and either the collective effective dose committed by one year of performance of the practice is no more than about 1 man·Sv or an assessment for the optimization of protection shows that exemption is the optimum option; and (ii) a series of limiting (bounding) use and disposal scenarios. The values of the activity concentration and total activity represent the lowest values calculated in any scenario for a moderate quantity of material. (b) In the case of more than one radionuclide, the appropriate sum of the ratios of the activity or activity concentration shall be taken into account. (c) Unless the exposure is excluded, exemption for bulk amounts of materials with activity concentrations lower than the guidance exemption levels of Table XII may nevertheless require further consideration by the regulatory authority.

safety/control programmes implemented in many countries, the actual radiation dose to workers has decreased significantly in the past two decades.

The occupational exposure of any worker shall be controlled such that the following limits are not exceeded [1]:

- — An effective dose of 20 mSv per year averaged over five consecutive years,
- — An effective dose of 50 mSv in any single year,
- — An equivalent dose to the lens of the eye of 150 mSv in a year,
- — An equivalent dose to the extremities (hands and feet), or the skin of 500 mSv in a year.

In the case of internal dose, the annual limit on intake (ALI) is based on a committed effective dose of 20 mSv. ALI values for ingestion and inhalation routes [5] and the Derived Air Concentration (DAC) values for enforcement in operating areas are presented in Annex II.

To comply with specified dose limits, the sum of the personal equivalent dose from external exposure to penetrating radiation in the specified period and the committed equivalent dose or committed effective dose, as appropriate, from intakes of radioactive substances in the same period shall be used. The period for calculating the committed effective dose shall normally be 50 years for intake by adults. To control risk from occupational dose as well as to comply with specific dose limits, consideration should be given to the cost effectiveness of additional measures to both reduce normal doses and those potentially arising from accidents. These measures could include, for example, changes to plant processes, operational regimes or shielding. Both the frequency and potential consequences of accidents should be considered.

6.4. DOSE LIMIT TO THE PUBLIC

The estimated average doses to the relevant critical groups of members of the public that are attributable to practices shall not exceed the following limits [1]:

- An effective dose of 1 mSv in a year,
- In special circumstances, an effective dose of up to 5 mSv in a single year provided that the average dose over five consecutive years does not exceed 1 mSv per year,
- An equivalent dose to the lens of the eye of 15 mSv in a year,
- An equivalent dose to the skin of 50 mSv in a year.

The three pathways that contribute to the radiation dose are the air, water and terrestrial routes and thus the annual dose limit of 1 mSv applies to the combined contribution from these routes. Discharge limits for the radionuclides for the air and water routes are established on this basis. The contribution from the terrestrial route arises from the disposal of radioactive waste in near surface and deep disposal facilities and depends on the half-lives and the activities of the radionuclides. These disposal facilities are designed to maintain their integrity for very long periods. Typically, during the service life of the nuclear facility, doses to the public through the terrestrial route are insignificant.

If the nuclear facility under consideration is one amongst many other nuclear facilities at a site, the annual dose limit of 1 mSv should be apportioned amongst all the facilities at the site, with due consideration for the nature, quantity and radiotoxicity of the nuclides discharged to the environment from each facility. For example, in the case of a MOX fuel fabrication facility with stringent containment and efficient ventilation systems, the discharge of alpha activity under normal operation through the air route will make an insignificant contribution to the dose received by members of the public. Similarly, liquid waste discharged from the plant, arising from decontamination operations, washings, showers, etc., may not carry any significant activity. However, it may be prudent to collect the liquid waste in a hold-up tank/sump for monitoring prior to discharge to the environment within the authorized limits.

Radiation doses to the public in the event of accidental release from the plant shall be managed according to the off-site emergency plans drawn up by the plant and local civil authorities.

6.5. OPTIMIZATION OF PROTECTION (ALARA)

6.5.1. General approach

The ICRP has recommended a philosophy of radiation protection based upon quantitative risk. This has been adopted in the new BSS [1]. The basic system was set forth in ICRP 26 [2]:

> “Most decisions about human activities are based on an implicit form of balancing of costs and benefits leading to the conclusion that the conduct of a chosen practice is worthwhile. Less generally, it is also recognized that the conduct of the chosen practice should be adjusted to maximize the benefit to the individual or to society. In radiation protection, it is becoming possible to formalize these broad decision-making procedures, though not always to quantify them.
> The Commission recommends a system of dose limitation, the main features of which are as follows:
>
> (a) No practice shall be adopted unless its introduction produces a positive net benefit;
> (b) All exposures shall be kept as low as reasonably achievable, economic and social factors being taken into account;
> (c) The dose equivalent to individuals shall not exceed the limits recommended for the appropriate circumstances by the Commission.”

The ICRP has stated that its recommendation that radiation be as low as reasonably achievable (ALARA) is synonymous with a recommendation for “optimization of radiation protection”. Because the ICRP system is based upon quantitative risk and detriment, it facilitates a quantitative description of the ALARA (optimization) recommendation based upon cost–benefit considerations. This approach was explored in considerable detail in ICRP 37 [6]. This report recognizes that quantitative techniques have both strengths and weaknesses [6]:

> "Optimization of radiation protection applies to all situations where radiation exposures can be controlled by protection measures... The degree of quantification in the techniques used for radiation protection optimization will vary with the different applications. Designers of installations and protection systems will tend to use more quantitative techniques for deciding the degree of protection (shielding thickness, containment, ventilation rates, etc.) that will meet the optimization requirement. Competent authorities may use stylized quantitative techniques of optimization in deriving appropriate authorized limits and requirements for given types of installations, radiation sources or practices involving radiation exposures."
>
> "Optimization of radiation protection during operations usually, but not necessarily always, tends to be less quantitative... Quantitative assessments of radiation protection optimization are not suggested for daily operational practice. The persons responsible for radiation protection in daily operations will have to follow simple rules imposed by the competent authority or the management, on the basis of the optimization principle. In addition, they may be guided by the general ambition of optimizing radiation protection, although in an intuitive rather than quantitative way."

6.5.2 Optimization of protection in public exposure

In practice, almost all public exposure is controlled by the procedures of constrained optimization and the use of prescriptive limits. It is often convenient to classify together individuals who form a homogeneous group with respect to their exposures to a single source. When such a group is typical of those most highly exposed by that source, it is known as a critical group. The dose constraint should be applied to the mean dose in the critical group from the source for which the protection is being optimized. Occasionally, the same group will also be critical for other sources, or, if the critical groups are different, each group may incur some dose from the sources for which it is non-critical. If the exposures in any critical group are likely to approach the dose limit for public exposure (see Section 6.4), the constraints applied to each source must be selected to allow for any significant contribution from other sources to the exposure of the critical group. The main aim of constrained optimization in public exposure should be to develop practical restrictions of the sources of exposure, e.g. in the form of restrictions on the release of radioactive waste to the environment.

6.6. REGULATORY GUIDES

Adequate safety provisions in the design (Section 7), adherence to written and approved operating procedures, regular monitoring of the work places (Section 8) and personal monitoring of the workers are prime requirements for all plutonium handling operations. A number of regulatory guides have been issued by the United States Nuclear Regulatory Commission (NRC) [7–23] on important safety features related to plutonium processing and fuel fabrication plants such as seismic design classification, quality assurance programme requirements, design of ventilation systems, design of liquid waste treatment systems and fire protection aspects. In the area of criticality safety, the American National Standards Institute (ANSI) has issued a number of standards [24–34] that deal with such subjects as criticality safety in operations with fissionable materials, plutonium–uranium fuel mixtures outside reactors, the storage of fissile materials, operations where shielding protects personnel, requirements for the criticality accident alarm system and assumptions for evaluating the radiological consequences of accidental criticality. In the USA, standards are prescriptive. In other countries, such as the United Kingdom, operators must demonstrate a level of safety which exceeds minimum standards.

6.7. CRITICALITY SAFETY LIMITS

In formulating safe operating limits to prevent criticality, all pertinent process conditions and failure possibilities must be taken into consideration. Specified limits may be derived from experimental data, wherever applicable. The limits based on theoretical calculations may also be specified, provided the calculational model is validated against experimental data. Safety factors [35] must be included in all limits and shall be appropriate for the degree of risk involved. Limits for the storage of plutonium and its compounds must also be based on experimental data or the results of validated computational techniques. Subcriticality of the store shall be ensured under all foreseeable conditions, including flood, fire, earthquake or other natural calamities. The relevant criticality safety data are given in Annex III.

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7. SAFETY ASPECTS OF DESIGN

The principal hazards associated with the handling and storage of plutonium are the consequences of its physical, chemical and radiological properties, and include contamination, radiation dose, fire and criticality accidents. Both nuclear and other non-nuclear hazards are taken into account in the safety requirements and in the design and operation of any plutonium facility. The safety related design principles for each type of hazard are developed in this section [1, 2]. Some concrete examples of the application of these concepts are given in Annex I.

7.1. SAFETY ASSESSMENT

A nuclear facility is considered to be safe if the provisions made during its design, construction, operation and decommissioning ensure that:

- Safety is maintained during normal operations;
- Any excursion outside normal operating parameters can be avoided;
- If an excursion outside normal operating parameters occurs, its effects can be effectively controlled.

The safety assessment of nuclear facilities is based upon three complementary approaches:

- Compliance with safety principles related to design and operation, as presented in Sections 7.2–7.14;
- Feedback of operating experience, examples of which are given in Section 7.15;
- Analysis of the potential risks based on:

    (1) identification of hazard scenarios and consequences,
    (2) estimation of the frequency of occurrence of the identified hazard scenarios.

Central to this approach is the concept of defence in depth, which requires successive barriers to prevent the release of radioactive material to the environment. Analysis of a postulated accident is normally based on a set of conservative assumptions to show that specific acceptance criteria have been met.

A number of techniques are available to conduct the safety assessment, for example fault tree analysis, event tree analysis, failure mode analysis, hazard and operability studies and probabilistic safety analysis [3]. These techniques depend upon a rigorous analytical approach by a person or teams with experience in safety assessment and plutonium facility operations. The output of such analyses permits comparison with pre-established criteria and, if necessary, modifications to ensure compliance with the criteria. This approach provides confidence that all major hazards from both normal and abnormal operation have been identified. Hazards and operability studies provide a comprehensive assessment of operational hazards. Once identified, hazard analysis can be used to quantify the risk, thereby allowing numerical comparison with pre-established criteria. When reliability of data allows sufficient accuracy, probabilistic safety analysis may be used.

During the safety assessment of a facility, human factors (i.e. how personnel behave and perform), must also be considered. The ergonomic aspects of the design will affect normal operations in areas such as radiation exposures, fatigue and identification of plant excursions, and will have an impact on the frequency and consequences of misoperations. The response to an abnormal situation should be considered both in terms of timeliness and probability of the correct action being taken to mitigate the consequences of the event.

7.2. CONTAINMENT

Plutonium containment prevents the spread of radioactive material to workers and controls its release to keep as low as reasonably achievable the quantity of radioactive material dispersed into operating facilities and the surrounding environment. To ensure containment integrity, facilities and equipment are designed to avoid fire and explosion, remove heat, and prevent mechanical failure (including drop of load) that may threaten containment integrity.

Plutonium facilities should be divided into a number of operating zones or areas by using barriers to separate areas of different risk from radiation and contamination. In practice, containment is achieved by barriers (static containment) and air flow using ventilation systems to establish pressure gradients (dynamic containment). The dynamic containment systems include air filtering units. In addition, monitoring is carried out continuously to detect any failure of containment (see Section 8).

7.2.1. Static containment

Static containment is normally provided by three physical barriers between plutonium and the environment (see Fig. 4). Table XII includes some examples of static containment methods. However, static containment is not sufficient in all circumstances.

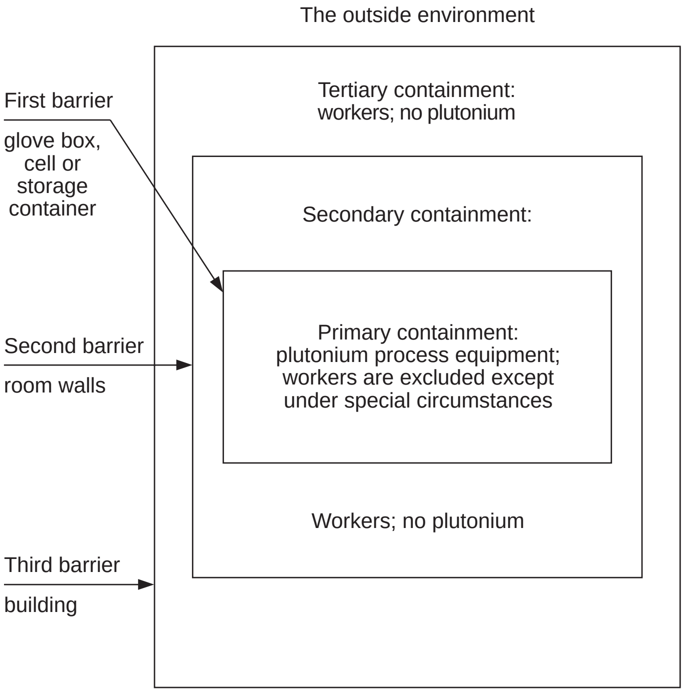

FIG. 4. Schematic drawing of three-physical-barrier static containment.

7.2.1.1. Primary containment

Primary containment is the barrier between the worker and plutonium. For solid plutonium, such as oxide powder, the primary containment which prevents plutonium from being released to the room is generally a glove box, cell or a storage container, as well as associated equipment such as an off-gas treatment system, if any. If a storage container is in a glove box, the glove box is then considered to be the primary containment.

For liquids and industrial applications such as MOX fabrication, essentially all of the plutonium is contained in the process equipment which is maintained at a negative pressure of about 70 mm WG (water gauge). Containment of all the contamination between the operation and the workers is either by means of the glove box

TABLE XII. STATIC CONTAINMENT METHODS

Class	Material form		
	Gas	Liquid and (whenever possible) solid	Solid
Primary containment	Equipment, piping, vessel, vessel ventilation system	Equipment, piping vessel	Glove box, storage containers, fuel cladding
Secondary containment	Cell, cell ventilation system	Cell, cell lining, or glove box	Room
Tertiary containment	Building, building ventilation system	Building	Building

or the cell maintained at a negative pressure of about 25 mm WG[1]. The primary containment may also consist of process equipment (piping, vessels, etc.) as well as associated equipment such as an off-gas treatment system — if they serve as the contamination barrier between the worker and the plutonium.

7.2.1.2. Secondary containment

The secondary containment includes the room walls and associated systems (ventilation, drip trays, etc.). Workers are permitted within the zone between the primary and secondary barriers. Plutonium is permitted in this zone only if it is totally contained in a transfer or storage container.

7.2.1.3. Tertiary containment

Tertiary containment includes the building and the associated systems and is complemented by the dynamic containment provided by the building ventilation system (dynamic containment is described in the next section).

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[1] The glove box is typically maintained at a negative pressure of 25 mm WG because a higher negative pressure would reduce the flexibility and therefore the usefulness of the gloves in the glove box.

7.2.2. Dynamic containment

Dynamic containment complements static containment. It is based on a series of negative pressure differentials. The system is designed so that the pressure is lowest in the areas where the plutonium is contained (process equipment and glove boxes), so that if a leak occurs, the flow is from the low contamination toward the high contamination area. The dynamic containment is provided primarily by the ventilation system, which is described in Section 7.3. The building ventilation network is connected to the final filter units, which include at least two HEPA filters at the exhaust to avoid the release of radioactive material to the surrounding environment.

7.2.3. Discontinuity of primary containment

A discontinuity of primary containment may be permitted for operational purposes. Piping emerging from process equipment constitutes an extension of the primary containment up to the first containment device such as a filter, a hydraulic seal or a valve. This applies, for example, to compressed air, steam, vacuum, nitrogen, ventilation air and liquid reagent piping. Process equipment that has moving parts also presents primary containment discontinuities. In such equipment, radioactive material containment is maintained by auxiliary devices such as seals, casing and covers, hydraulic seals and air current directed into the equipment. A discontinuity of static containment may also be the result of an accident (see Sections 7.4, 7.5 and 7.8).

Allowance must be made for carrying out certain operations requiring the temporary removal of the barrier, such as:

- Introducing material into process equipment and removing products and by-products during operation,
- Replacing equipment/components.

In order to prevent the spread of radioactive material during these operations:

- Temporary containment is installed where the containment discontinuity is to occur; for example, a plastic tent may be erected, or a ‘bag out’ device may be installed;
- Containment continuity should be restored as soon as possible;
- Where primary containment must be penetrated to remove or replace worn-out parts of equipment components, the following measures may be taken:
    (1) Provide dynamic containment by the use of suitably directed air currents, or by an air curtain;
    (2) Decontamination of worn out parts before removing.

- Special maintenance devices, such as a cask with a double containment door, for equipment (pumps, valves, etc.) may be used.

7.2.4. Containment monitoring

At least one static containment barrier (normally primary containment) must be provided between the plutonium source and the workers. And at least one of the barriers between the plutonium and the workers must be monitored to detect any leakage of radioactive material. The principal means provided for this surveillance (which are discussed in Section 8) are:

- Checking the air from the space adjacent to the first barrier, for instance the air in rooms which contain plutonium handling glove boxes is continuously monitored by air sampling and alpha detectors. Possible surface contamination is also checked periodically.
- Continuous monitoring of the gaseous effluents.

For liquid processes specifically:

- A high level alarm and a means to take samples in the drip tray sumps,
- Introducing fibrescopes and cameras into cells,
- Flushing the drip trays and verifying (by sampling) the level of contamination after flushing,
- Checking the heat transfer fluids for plutonium contamination.

The monitoring of the containment is mainly carried out using radiation monitoring devices. Before release to the environment through the stacks, the amount of radioactive material in the gas is monitored. Liquid radioactive effluent is also monitored for activity content before release (Section 7.13.1).

7.3. VENTILATION

Nuclear facilities are provided with ventilation systems ensuring several safety functions:

- Dynamic containment, as defined in Section 7.2.2. Ventilation systems move air from uncontaminated zones accessible to staff to zones containing process equipment.
- Sufficient air changes in zones accessible to personnel.
- Treatment of off-gases from the process systems and the building to remove possible contamination.
- Containment monitoring (see Section 7.2.4).

Ventilation systems, including supply and extraction fans, create a series of negative pressure gradients, the pressure being set at a value related to the potential hazard of the materials contained. This results in grouping in the same zone of a facility of materials of similar hazards, with the central zone of containment housing the process equipment and creating a series of negative pressure gradients from the peripheral zone to the central zone. Leakage of air through the containment is thus, in normal operation, towards the central zone, which prevents the spread of radioactive materials either into occupied zones or into the environment.

The required quality and reliability of the ventilation system (especially the redundancy of the components and the electrical power supply) is determined by the consequences that failure could cause for workers and the environment. To limit the exposure of workers maintaining the ventilation system, and to maintain the facility in a state of radiological cleanness, ventilation systems must be of high quality. They must also be specially designed so that they do not constitute weak points during abnormal (though rare) conditions such as fire.

7.3.1. Process equipment ventilation

Process equipment ventilation is distinctly separated from the glove box, room and building ventilation.

- The process equipment ventilation system includes:

  (1) Collection of off-gas from process equipment,
  (2) Treatment of off-gas (process unit),
  (3) Extraction and filtering of off-gas by fans and subsequent release through the stack.

- Off-gas exhausted from all process equipment containing radioactive liquids or solids must be treated (decontaminated) before being released to the surrounding environment;
- Treatment is selected according to the concentration level of the chemical or radioactive material involved;
- The off-gas networks are designed to minimize the risks of contamination transfer from equipment.

Specifically, this ventilation system has:

- Two filtering stages (HEPA filters) in service,
- Provisions to prevent, as far as possible, excess pressures in process equipment,
- Provision to send gases to the stack after filtering and monitoring.

7.3.2. Glove box and cell ventilation system

Glove boxes are maintained at a suitable negative pressure relative to the room in which they are located to ensure a sufficient air flow rate in case of a containment breach. The usual pressure of the glove box is about 25 to 30 mm WG less than that in the surrounding room (see Figs 5 and 6). Off-gases from the glove boxes are usually filtered by at least three HEPA filters in series before discharge to the environment. The first stage is typically located near the glove box and serves to minimize plutonium contamination in the ventilation lines. Because of the HEPA filter's proximity to the glove box, it is often not testable, and therefore no credit is given for it in the safety analyses.

7.3.3. Building ventilation systems

The main purpose of the building ventilation system is to maintain the cells and glove boxes at a lower pressure than the rooms in which they are located in order to ensure dynamic containment and to draw radioactive material towards the filters. The number of filters and the design of each ventilation network is linked to the expected contamination level in each room to prevent accumulations of radioactive material in ducts. Figure 5 shows a schematic diagram of the organization of ventilation, while Fig. 6 shows an example of the pressure profile in a plutonium handling facility.

7.4. FIRE SAFETY

Fire hazard in a plutonium facility results from:

- Sparking from electrical and electronic equipment,
- The utilization and storage of flammable liquids (solvents, diluents, etc.) related to the process,
- The presence, use and storage of pyrophoric materials, especially plutonium and non-stoichiometric plutonium oxide in air.

7.4.1. Analysis and methodology

Fire safety begins with the identification of all credible situations in which a nuclear risk might arise as a result of a fire. The next step is to identify the effects for each credible situation. All reasonable measures are then adopted to minimize the likelihood, severity and consequences of potential fires. The techniques described in Section 7.1 are used to carry out this analysis. The event tree method may be used to determine possible fire scenarios and the most suitable means to prevent, detect and extinguish them.

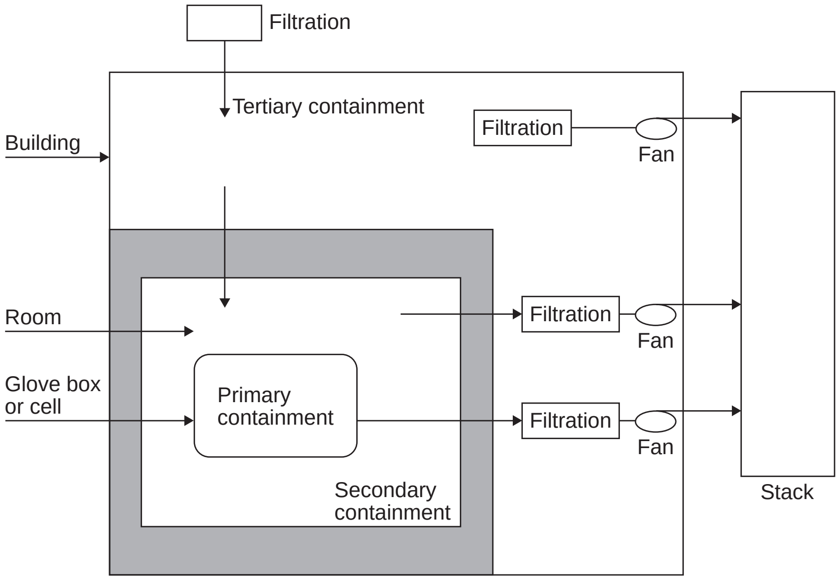

FIG. 5. Schematic diagram of a ventilation system.

Preventive measures are essential: they are determined at the design stage, on the basis of the fire hazards arising from outside the facility, from the processes utilized and from the operating conditions inside the facility. Detection devices must be arranged so that the outbreak of fire can be detected at all times. Means must also be provided for limiting the spread of fire and for extinguishing it.

7.4.2. Fire prevention

Fire prevention can be considered in three categories.

— Controlling ignition sources:

(1) An inert gas (i.e. nitrogen or argon) is often used to avoid the risk of igniting gases, flammable liquids and pyrophoric solids.
(2) Grounding of equipment with static electricity risks.
(3) Where flammable reagents are necessary, the process is maintained outside the flammable region of the reagents.

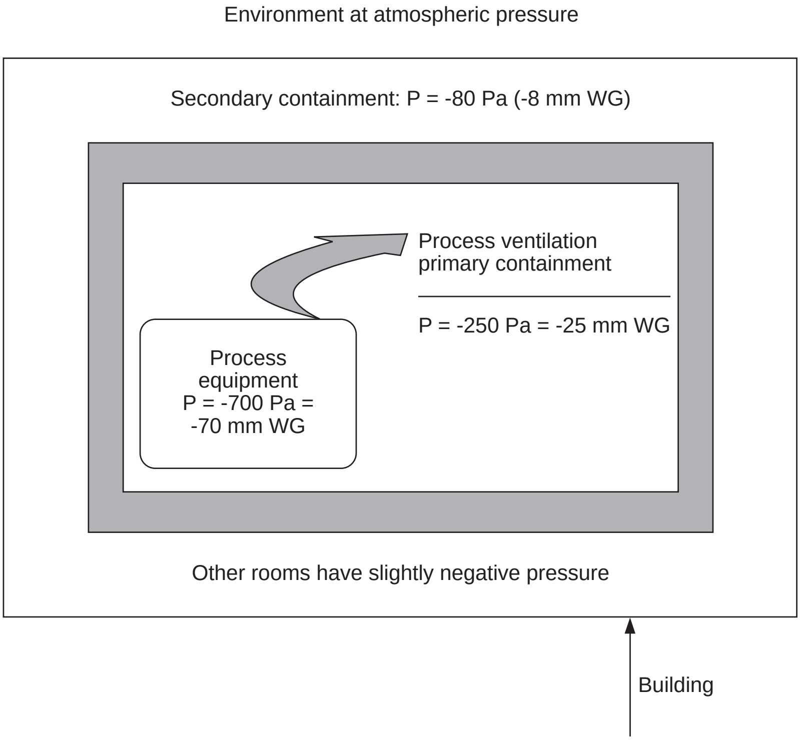

FIG. 6. Typical plutonium facility pressure profile (WG: water gauge).

(4) Electrical equipment should be designed to avoid fire risks and should be installed outside the primary containment if possible. For example, except in glove boxes where an inert atmosphere is used, electric motors should be mounted outside the primary containment or, where this is not possible, a motor of flameproof design must be used.

— Controlling and minimizing the use of combustible materials:

(1) The fire load per unit area in the rooms may be limited by the selection of suitable construction materials and equipment on the basis of their flammability and minimization of combustible materials in the rooms; if possible the process equipment is contained in separate cells or glove boxes of relatively small dimensions.
(2) Flame retarding electrical cables can be used.

— Reducing inventory involved in a fire:

(1) Based on an analysis of fire risks, rooms and cells may be divided into several fire classes according to their calorific load;
(2) Physical separation is maintained between zones where there is a fire hazard and the remainder of the facility;
(3) Physical separation is maintained between zones containing equipment important for safety and the remainder of the facility.

Physical separation, which is to prevent the spread of fire, is provided by the fire resistance of the boundaries of the zones. These zones are designated ‘fire zones’. The fire resistance required for the boundaries (including the doors, utility ducts and ventilation ducts) is determined on the basis of the probable duration of the worst fire which could affect each boundary, with allowance made for the fire load of the rooms. The zones containing quantities of radioactive materials that would give rise to unacceptable consequences in the case of a fire are specially designed. The containment systems for these zones consider the possibility of fire and ensure adequate capability to filter the hot gases extracted from the zones in the event of fire. The ventilation network can be arranged so as to provide at least one filtration level with additional protection against fire by diluting the hot air from the room on fire by cool air from rooms unaffected by the fire.

In addition to the general provisions mentioned above, preventive measures must be used in rooms containing flammable materials, and which are accessible to staff. These measures include the use of explosion proof electrical equipment, safety type electric light fixtures, and the installation of guards, casings or screens to control any leaks of flammable liquid.

7.4.3. Fire detection

In all rooms where the risk of fire exists, an early detection system, often associated with an automatic extinguishing system, should be provided. The information transmitted by this system should be displayed in the facility control room. The rooms containing safety equipment must also be protected by a fire barrier. Corridors must be protected by a fire barrier to enable evacuation of workers and access by the fire brigade.

7.4.4. Extinguishing the fire

The strategy for fire fighting inside the plutonium plant centres around the containment barriers. Provisions for fire fighting must be available and must be of a suitable nature for controlling fires in a period of time compatible with the fire resistance of the containment system (boundaries, corridors, ventilation ducts and filters) before significant radiological consequences ensue. Therefore, the design and operation of the ventilation systems play an essential role. For instance, whenever possible an air extraction filtration stage is provided downstream of the zone where the extraction ducts of the different zones join to permit the dilution of the hot air from one fire zone by the air from unaffected zones. Furthermore, it may be possible, if needed, to isolate the air supply system of a zone with a fire damper.

The means of extinguishment may be fixed or mobile. Fixed equipment (for instance systems for spraying water or dispensing extinguishing gases or carbon dioxide) are used where the access is difficult. Since water is an effective moderator and will decrease the amount of plutonium required for criticality, the criticality hazard must be examined when water is considered as the extinguishing agent in a zone containing fissile materials. If such a hazard exists, the use of water must be prohibited and non-hydrogenated extinguishing agents must be used instead. Whatever extinguishing agent is used, consideration must be given to the potential change in geometry which may result from its use. A means to retain potentially contaminated liquid extinguishing agents is normally provided. In rooms where the fire hazard is not accompanied by a radiological risk, the fire fighting action is conventional. The various arrangements to address the fire hazard must also ensure avoidance of unacceptable radiological consequences.

7.5. PREVENTION OF EXPLOSIONS

In a plutonium facility the main risk of explosion is linked to the use of solvents and diluents, to the use of hydrazine and hydrogen and to the generation of hydrogen by radiolysis. Since nitrate and some solvents, like TBP, can react under specific conditions, precautions must be taken to avoid entrainment of an organic phase in the evaporators. The temperature of the heating fluid is also limited to avoid the domain where TBP and nitrate could react. Temperature in the process is maintained below the flashpoint of the diluent or solvent and spark proof electric appliances are used when a diluent or solvent is present. The reagent vessel containing highly concentrated hydrazine is purged by nitrogen. The concentration of hydrazine decomposition products is controlled.

To prevent explosions caused by the accumulation of hydrogen generated by radiolysis, highly reliable systems are used to dilute the tank atmospheres with air or nitrogen. In furnaces operated with hydrogen atmosphere, hydrogen gas is generally diluted by inert gas (nitrogen or argon) below its explosive limit and the hydrogen concentration is monitored continuously.

To deal with risks of explosion of external origin related to industrial activity or to the transportation of dangerous materials, the risks are analysed to determine if special reinforcement is required. Moreover, the siting of the nuclear facility itself should be such that the facility is located far away from the potential sources of an explosion.

7.6. CRITICALITY SAFETY

The criticality risk is analysed early in the design of plants, since criticality prevention measures will affect the choice, design and layout of equipment. Criticality safety parameters are given in Annex III.

7.6.1. Methods to control criticality

Criticality can be affected by mass, shape, volume, moderation interaction, neutron absorption, reflection and density. Generally, criticality is avoided by using one or more of the following five methods:

- Restricted geometry,
- Mass and moderation control,
- Limitation of concentration of fissile materials,
- Neutron absorbers,
- Spacing of equipment.

7.6.1.1. Restricted geometry

The dimensions and shape of items of equipment may be chosen so that the fissile media remain subcritical for a given physical or chemical form of the fissile materials. The configuration is then said to be of ‘safe geometry’. An example of this criticality control method is to limit the diameter of a pulsed column used for the extraction of plutonium by a solvent.

This method of geometry control necessitates a number of precautions:

- Maintaining the geometry despite an initiating event liable to cause deformation (earthquakes, dropped loads, fires, etc.);
- Choosing, for corrosive fissile solutions, equipment of high corrosion resistance to avoid leakage, and providing means of detecting and recovering leaked material;
- Analysing the situations created by the possible transfer of fissile solutions from equipment of safe geometry to equipment of unsafe geometry, or by inadvertently locating fissile materials near the ‘geometrically safe’ equipment, and ensuring that such situations are avoided.

The restricted geometry method is the most appropriate for industrial facilities using continuous processes.

7.6.1.2. Mass and moderation control

The mass of fissile matter in the functional unit is limited to less than the critical mass (corresponding to a neutron multiplication factor = 1). This mass depends on the moderation ratio (the ratio between the number of equivalent hydrogen atoms and the number of atoms of fissile elements), the specific mass of fissile material and the neutron reflection conditions. Control of criticality risk by limiting the mass of fissile materials involves careful accounting of these materials in each functional unit, with allowance for uncertainties.

Setting the upper mass limit below the critical mass corresponding to the optimum moderation ratio makes it unnecessary to control the moderation ratio. When such a limit is incompatible with the process utilized, a higher mass limit may be used provided the domain of the moderation ratio is controlled and arrangements are made to adhere to it (e.g. the use of water in the event of a fire may be forbidden where the moderation ratio must be maintained within a specified domain). When mass control is used in a low moderation region, such as in plutonium dioxide handling operations, the moderating conditions shall be determined considering the lubricant, organic binder and moisture in the oxide.

7.6.1.3. Limitation of the concentration of fissile materials

This method is limited to homogeneous solutions of fissile materials. It requires both continuous information on concentration and precautions to avoid the accumulation of fissile materials by precipitation and deposition, or by increasing the concentration (e.g. by evaporation due to heat). This method is applied to processes in which the concentration is far below the critical limit in both normal operations and in the case of an incident.

7.6.1.4. Neutron absorbers

Neutron absorbers with a high neutron capture cross-section can be used to avoid criticality. Neutron absorbers can be either soluble in solution with plutonium (e.g. gadolinium nitrate) or solid placed inside (for instance borosilicate rasching rings), or around the vessel containing plutonium (such as borated concrete). This control method assumes that the concentration of the absorber elements is maintained within a specified range in the liquid and the absorbing solid remains intact under chemical attack or in the event of an incident (earthquake, drop load, fire, chemical attack, etc.).

7.6.1.5. Spacing of equipment

Systems that are subcritical individually may become critical if they are put too close together. Adequate spacing between the subcritical systems must be calculated on a case by case basis under the worst moderation and reflection conditions (e.g. flooding or the presence of mists) in the design of the plant, in the design of the storage facilities and in the transport of the fissile material. The maintenance of the spacing under all conditions has to be ensured. Introduction of a hydrogenous material (e.g. polyethylene or concrete) of suitable thickness between the subcritical systems helps to reduce the spacing between them and thus conserve valuable space in the process area. If the isolating hydrogenous material is encased in a cadmium sheet or is homogeneously mixed with neutron absorbers (e.g. borated polyethylene or borated concrete), its required thickness can be reduced and the equipment spacing can be reduced further.

The above methods of criticality control may be used alone or in combination, depending on their compatibility with the operating constraints of the processes. In practice, the possibility of criticality in each unit is evaluated and prevented, taking into account the:

- Composition of the fissile material,
- Physical and chemical properties of the fissile material,
- Presence of a neutron moderator or reflectors,
- Potential for neutron interaction between units.

For each functional unit that represents a criticality risk, a reference configuration is established: shapes and dimensions of materials, composition of materials in terms of fissile and non-fissile atoms and the conditions of reflection of neutrons at the boundaries. Criticality studies are carried out for each functional unit of a nuclear facility and include safety margins (see Annex III–3) resulting from the use of conservative estimates. In addition, the upper limit for the K_{eff} calculation, which includes computational errors and uncertainties always set lower than 1 — typically 0.95 is used.

7.6.2. Double contingency principle

The design arrangement selected for a plant should also prevent a criticality accident. The following principle is used as the basis for the analysis of criticality hazard:

- A single anomaly such as a component failure, a function failure, a human error (e.g. instruction not followed), an accident situation (fire for instance), must not result in a criticality accident.

— If a criticality accident can occur because of the simultaneous occurrence of two anomalies, it must be shown that:

(1) The two anomalies are strictly independent (not common mode),
(2) The probability of occurrence of each anomaly is below some threshold,
(3) Each anomaly can be detected with suitable and reliable means within a time that allows countermeasures to be taken.

7.6.3. Mitigating the consequences of a critical excursion

The preventive measures derived from the principle described above make a criticality accident highly improbable. However, the possibility of such an accident must be considered and its consequences studied to assess the need for mitigative measures, and especially to ensure that workers are protected.

7.6.4. Criticality detection and alarm devices

Detection and alarm devices are used to alert personnel, and to permit rapid evacuation to protect the staff. These are required in the zones accessible to personnel which may experience a significant increase in the radiation level in the case of a criticality accident. The criticality detection and alarm system triggers the evacuation of personnel. It could also permit the radiation dose rate to be measured to provide guidance in any rescue operation. Typical radiation exposures from a criticality accident of 10^{18} fissions are provided in Table II–5 and in Figs II–2 and II–3 in Annex II.

7.6.5. Environmental consequences of a typical criticality excursion

The calculation of the releases of radioactive substances and of external doses was carried out for a reprocessing plant in which a criticality resulted in 5×10^{18} fissions in 10 min. From this calculation, the paths for evacuation of the operators from the radiation field were determined. The consequences to the environment of a criticality excursion of 5×10^{18} fissions have been evaluated with the assumption that the fission products released are entrained by the process ventilation and are trapped in the filters — except halogens and rare gases which are discharged through the stack. For the halogens, this is a conservative assumption (the criticality experiments performed by the French Institut de protection et de sûreté nucléaire show that 10% or less of iodine produced in the course of the criticality excursion would be released into the environment).

7.7. REMOVAL OF DECAY HEAT

Heat generation from plutonium from LWR fuels is in the range of 10–25 W/kg, as discussed in Section 3. Therefore, consideration of decay heat is necessary for the handling and storage of large amounts of plutonium. Cooling is usually provided for solutions with a high concentration of plutonium by circulation of a cooling liquid and for cooling solid plutonium by circulating air.

7.8. PREVENTION OF MECHANICAL FAILURES OR LOAD DROP

Containment could be breached by a mechanical failure if a load is dropped on a vessel containing plutonium, or a container of plutonium is dropped. This risk is minimized by designing material handling equipment and lifting gear in such a way as to reduce the probability of such accidents to a very small value and by providing floors with sufficient mechanical strength to support them. The maintenance and periodic inspection of cranes and manipulators should be considered in facility design.

7.9. RADIATION SHIELDING

Radiation exposure of personnel must be less than the limits defined by regulation. Moreover, in compliance with the ALARA concept, measures should be taken to minimize this exposure. As discussed in Section 3, plutonium and its daughters emit alpha and beta particles, neutrons, and gamma rays. Only gamma rays and neutrons have sufficient penetrating power to require special attention when considering external exposure. For these sources, radiation shielding, automation and remote controlled operating systems are normally used. Alpha and beta radiation do not require additional shielding.

The gamma energy spectrum depends mainly on the plutonium isotopic composition and the presence of ^{241}Am produced by the beta decay of ^{241}Pu. In plutonium purification plants, process equipment is generally welded and located in concrete shielded cells. The thickness of the concrete wall for seismic safety often exceeds that for gamma ray protection. The gamma and neutron radiation from civil grade plutonium is much higher than for weapon grade material. Therefore, in the new facilities, plutonium is not routinely handled with gloves (with the exception, perhaps, for analysis carried out in laboratory facilities). Most of the industrial operations for civil plutonium are now automated and remotely controlled, but many plutonium operations in laboratories (particularly using plutonium with a high ^{239}Pu content) may be conducted manually in glove boxes. The presence of workers is required mainly for maintenance operations. Nevertheless, these operations are carried out after removal of most of the plutonium from the glove boxes. For neutron shielding the acrylic panels previously used are now often replaced by acrylic resins, polyethylene (or hydrogen rich material) and by materials with neutron absorbers such as boron. If the quantity of plutonium is small (or if shielding is provided within the glove box), acrylic panels can be used. For gamma ray shielding, concrete, lead compound acrylic resins, or other lead compound plastics are used.

7.10. COUNTERMEASURES FOR THE LOSS OF ELECTRICAL POWER

Plutonium facilities should be designed with an adequate and continuous electrical power supply, particularly to equipment such as ventilation fans, radiation monitors and fire alarms. To ensure such supply, redundant commercial electrical supply lines and emergency generators should be installed. Procedures should also define the restart sequence of ventilation fans to avoid back currents of exhaust gases.

7.11. SEISMIC SAFETY

7.11.1. Earthquake design basis requirements

All nuclear facilities, including those handling plutonium, are designed to withstand earthquakes [4]. The seismic safety requirements of each facility are defined in terms of the potential radiological consequence to the environment. The most severe earthquake assumed in the design of a facility is defined according to the local geographical conditions. In Europe, it is called the ‘safe shutdown earthquake’ (SSE).

Allowance for the SSE consists of adopting a design for the facilities such that the important safety functions continue after the earthquake’s occurrence. For that purpose, seismic classes are generally defined, taking into account the environmental impact caused by earthquake damage on the criticality safety, containment, fire and explosion safety, etc.

7.11.2. Radiological consequences of an SSE

An estimation of the radiological consequences of an SSE on the facility must be carried out and the facility design should minimize the radiological consequences. In general, the containment barrier (see Section 7.2) and the shields provide protection against exposure to radiation. The assurance that protection will be maintained against exposure to radiation is obtained by paraseismic civil engineering design.

7.12. AIRCRAFT CRASH HAZARD

The probability that a reference aircraft will crash into a given facility can be calculated. For example, analysis of the yearly probability of an aircraft crash causing unacceptable consequences at the French UP3-A plant is low enough to consider this hazard to be beyond the design basis. However, in the scope of beyond design basis accident studies, the tests carried out by the French Commissariat à l'énergie atomique demonstrated that 50 cm of concrete (which is representative of the facility walls) constitutes an effective shield against the impact of a business type aircraft.

7.13. MINIMIZATION OF RADIOACTIVE WASTE

7.13.1. Liquid waste

Liquid radioactive waste arises in the normal operation of a nuclear facility, including decontamination of equipment during maintenance operations. Arrangements are made to minimize the quantities and radioactivity of such liquid waste by recovering radioactive decontamination solutions and recycling them. No releases of liquid radioactive effluents into the subsoil are permitted. The equipment and tanks containing radioactive liquids must be designed to remain leak tight and must be equipped with leak recovery devices (drip trays). Radioactive effluents are released to the environment only after verification that their chemical and radioactive properties are within proper limits. All liquid effluent releases from a nuclear site must comply with the limits laid down in the license for liquid radioactive effluent release. Discharge to the environment is minimized through the appropriate management and treatment of liquid effluents. For instance, at the La Hague site, facilities handling plutonium have a waste management policy to produce either high level radioactive waste in limited volume (to be incorporated in glass for deep underground disposal), or low level radioactive effluent which can be directly discharged to the environment. The main features of the process are the use of evaporation to ensure high decontamination of the solution and the use of a salt-free process.

7.13.2. Solid waste

The radionuclides trapped in the liquid and gaseous effluent treatment processes constitute part of the solid waste produced by a facility. In addition, normal operation of a facility gives rise to 'technological waste' such as pieces of metal, glass or plastic with small amounts of radioactive substances on their surfaces. The solid waste produced is processed to immobilize the radioactive substances it contains to a form which is physically and chemically stable and of limited volume prior to interim storage, transport and final disposal. Three actions to limit the solid waste requiring disposal are:

- Reliable processes are used; miscellaneous waste, such as vinyl, is minimized;
- Solid waste is decontaminated to permit its reuse or to reduce the amount of radioactivity in the waste to be disposed;
- Wastes are compacted or incinerated where feasible.

The waste thus treated makes up ‘packages’, which comply with technical specifications concerning their radioactive content, their capability of confining the radionuclides they contain and their suitability for disposal in an appropriate permanent facility or for interim storage if a permanent facility is not available.

7.14. DESIGN FOR DECOMMISSIONING

Considerable experience has been gained by dismantling facilities which handled plutonium such as purification plants and radiometallurgical laboratories. This experience is used in designing and operating new facilities. For example, in a reprocessing facility:

- Equipment is laid out in separate cells or glove boxes according to its function and level of activity in order to facilitate access;
- Equipment is fitted with specific decontamination solution inlets and recirculation devices;
- Accumulation of radioactive material in the plant during normal operations can be avoided by carrying out a periodic flushing campaign (once or twice a year) to make decommissioning easier. The degree of decontamination achieved during such operation is usually sufficient to allow the maintenance of equipment.

These provisions allow the removal of much of the residual activity prior to dismantling in order to limit the contamination and to minimize the final waste to be stored in deep underground storage.

7.15. FEEDBACK FOR SAFETY IMPROVEMENTS

Considerable effort has been made to incorporate the experience gained from the previous plants in the design of the new plants. Working groups have been set up (usually consisting of the contractors, the operators, the sponsors of the process and the engineers) to consider process safety. Improvements in the safety provisions of new reprocessing plants and fuel fabrication plants result from the high safety standards set for these plants. These improvements include the use of advanced technology in reprocessing and fuel fabrication fields involving the use of computers, electronics, new maintenance concepts and advanced mechanization.

Feedback from safety experience has produced the following improvements:

- Modern facilities are operated remotely from a control room which allows supervision of all the events occurring in the plant and permits remedial actions as required by the situation. Some locally controlled operations still exist in areas having low radiation levels (such as preparation of new packages for waste).
- Sampling during normal operation is carried out using automatic devices, with no break in the containment while maintaining personnel protection against radiation.
- The engineered systems for containment of radioactive materials and ventilation networks are improved by the inclusion of:

  (a) more rigorous continuity of primary containment,
  (b) independent ventilation systems for the primary and secondary containment.

- In the packaging of plutonium oxide, aluminium containers enveloped in vinyl polychloride are replaced by more reliable welded stainless steel containers.
- More effective radiological surveillance devices are used.
- Maintenance operations are carried out during normal conditions without exceeding the exposure limits of the zone involved owing to:

  (1) Thorough preparation of procedures for maintenance and repair in the radioactive environment.
  (2) Modular design, which permits defective equipment (pumps, valves, stirrers of mixer–settlers, shielded filters, parts of sampling benches, ejector nozzles, etc.) and waste removal bins to be replaced without loss of containment and when required with protection against radiation by using mobile equipment replacement casks (MERC).
  (3) The use of simple equipment minimizing moving parts.
  (4) Locating motors outside the primary containment whenever possible.

The reduced average exposure of operating staff and reduced releases of liquid and gaseous effluents demonstrate the effectiveness of this safety feedback. Significant improvements have been made in the design, construction and operation of new reprocessing and fuel fabrication plants. The feedback of experience will continue to be used to further improve the safety of operation.

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8. OPERATIONAL SAFETY

The design considerations for plutonium handling facilities are discussed in Section 7, with examples of the implementation of these design features described in Annex I. The behaviour and consequences of an accidental release of activity to the environment and to personnel are described in Sections 4 and 5. This section describes the key features in managing a plant to ensure that it remains safe during operation, maintenance and modification. These features are:

- Effective management,
- Characterization during commissioning,
- Established operating procedures,
- Training,
- Control of nuclear material,
- Control of maintenance,
- Control of modifications,
- Audit of operations.

Radiation monitoring and emergency procedures are emphasized here.

8.1. EFFECTIVE MANAGEMENT

Effective and unambiguous control is essential both to ensure that current procedures reflect any learning experiences and that personnel carry out operations in a consistent manner that has been demonstrated to be safe [1]. The potential hazards presented by routine and non-routine operations should be assessed to ensure that adequately engineered (physical) and administrative (procedural) controls are in place. Management is responsible for ensuring that satisfactory levels of safety staffing and expertise are available to support operations. It should carry out audits on compliance to ensure that training and communications are effective. The status of the plant during operation, maintenance and modification needs to be clearly known.

8.2. CHARACTERIZATION DURING COMMISSIONING

The commissioning of a plant is carried out according to pre-agreed schedules. During commissioning, the lifetime operation and plant decommissioning are characterized. This ensures not only safe operation, but also that new designs and procedures will work as expected [2].

8.3. OPERATING PROCEDURES

All nuclear facilities must obtain a license to operate, as described in Section 6. This license will include a number of conditions that must be met in order for the facility to operate. In addition, national law, international guidelines (such as the new BSS [3]) and international principles contain other requirements which must be met for operation. The safety assessments for a plant will outline the assumptions for safe operation. Management will interpret these requirements into procedures and instructions to ensure that the plant remains within established safe limits. These procedures must be controlled to ensure that they remain current. Procedures cover, for example:

- Process and equipment operations,
- Preventive and breakdown maintenance,
- In-service inspection and quality assurance,
- Waste management,
- Radiation protection,
- Safeguards,
- Physical protection,
- Fissile material control,
- Emergency response and preparedness for:
    1. Criticality accident,
    2. Release of radioactivity at the site,
    3. Building emergency (fire, flooding, earthquake, etc.).

8.4. TRAINING

To ensure safe operation it is necessary to develop a safety culture [4]. Each person must be made aware of the potential risks, how to prevent accidents and, if accidents occur, how to limit the consequences to workers, the public, the plant and the environment, as described in Sections 8.12 and 8.13 [5–9]. Operators must be trained in procedures and tested on their understanding before being allowed to take up operational duties [10]. Most modern plants now have simulators to assist in training. The responsible manager assigns, usually in writing, key roles that have an impact on safety only to suitably qualified and experienced people. Personnel are required to attend periodic refresher courses. Training records are audited by regulatory bodies.

8.5. CONTROL OF MAINTENANCE

Ease of maintenance should be designed into modern nuclear facilities to minimize the proximity of maintenance personnel to nuclear material and consequently minimize the dose to them. However, maintenance operations often entail higher radiation exposure than normal operations and so work must be carefully managed to keep the radiation dose at ALARA levels [11]. Dose minimization is achieved by following maintenance instructions that have been assessed for safety and efficiency and that are periodically reviewed to update them in the light of operating experience [12]. As in conventional plant safety, when the plant is taken out of service for maintenance it must be formally handed over and locked out, i.e. put in a state where personnel cannot erroneously restart operation.

Plutonium must be contained (e.g. in a storage container, a glove box or a cell) to prevent the working area from being contaminated. Monitoring of airborne activity provides feedback on the integrity of containment and alerts personnel to any rise in airborne activity. During modifications to containment, which may require the use of temporary containment such as blister boxes or tents, it is particularly important to provide local air monitors and alarms. More local ventilation may also be needed. Personnel may be required to use additional personal protection equipment (including self-contained breathing air sets, protective clothing, respirators, etc.), in addition to monitoring equipment, to ensure that the dose remains within prescribed limits for the operation.

Facility and equipment design should ensure that gamma and neutron radiation is either effectively shielded or operators are excluded by using remote operations. Additional shielding and dose monitoring may be required during maintenance and modifications as the normal protective measures may have to be bypassed to conduct the maintenance or modification.

Before formal handover back to operation, the operational management must be informed with documentation of any changes to the plant.

8.6. CONTROL OF MODIFICATIONS

Modifications to the plant must meet the same requirements as routine or periodic maintenance. Safety assessments, however, must be made to cover not only the operation but the effect of the proposed changes on the future operations of the plant, including any credible fault scenarios. If the potential hazards are perceived to have major consequences, then the licensing authority will be expected to endorse the changes before they are authorized. Formalized procedures for authorizing changes require updating of drawings and documents before formal handover of the plant back to operation.

8.7. AUDIT OF OPERATIONS

Audits of training, operation, maintenance and modification for compliance with approved procedures is carried out routinely by the authorizing bodies in addition to audits, reviews and inspections by management [13–14]. Customers may also carry out audits to ensure that operations, processes and standards are as agreed. Requirements for the retention of records related to safety, i.e. operational records, are usually stipulated in the license to operate.

8.8. RADIATION MONITORING

Persons who work in controlled areas should be given specific training in radiation protection for the work to be performed. This should include pertinent information concerning monitoring devices, radiation protection policies and current practices that are in effect within the establishment [15].

The basic principles of occupational radiation management are described in Refs [11, 15]. Additional material can be found in Refs [3, 16, 17].

The safety of workers is ensured through a combination of area and personnel monitoring. Area monitoring provides a general assessment of work area conditions and early indicators of potential problems; personnel monitoring provides individual assessments of potential dose. Area monitoring, conducted by professional radiological control technicians, includes measuring ambient dose rates from gamma and neutron radiation, routine contamination surveys on work surfaces and measurement and assessment of airborne radioactivity. Personnel monitoring includes determining external and internal dose in routine operations and accident conditions.

8.9. AREA MONITORING

A plant is divided into different zones, depending on the radiation levels and potential risk to workers and the environment [3]. These zones are demarcated and may require the crossing of physical barriers to alert personnel of the increased hazard and any additional personal protective equipment which may be required. On crossing back from these barriers, contamination monitoring using hand, foot and clothing contamination monitors is usually required. Additional controls, such as washing of hands, may also be required.

8.9.1. Ambient radiation monitoring

Surveys should be conducted at a frequency commensurate with the operations. That is, they should be conducted more frequently in areas where work is being conducted and the dose rates fluctuate significantly, and less frequently in areas with low occupancy and unchanging dose rates. Survey results should be posted so that they can be utilized by workers. Readouts from installed ambient radiation monitors are usually displayed in a control room which is at a safe distance from the processing area under criticality accident conditions.

Radiation surveys using handheld survey meters provide important information to workers and can contribute significantly to keeping personnel doses as low as reasonably achievable. The instruments used to perform these surveys must be calibrated and functional, and be appropriate for the type and level of radiation expected.

Ambient radiation monitoring is conducted using a combination of installed (stationary) radiation monitors and handheld survey meters. Typical survey meters for gamma radiation include ionization chambers, NaI (Tl) scintillation detectors and Geiger–Müller (GM) tubes. To monitor neutron radiation doses directly, BF_3 detectors are typically used, which enable a reasonably accurate measurement of neutron radiation dose equivalent rates with little dependence on neutron energy.

Criticality detectors are an essential element of the ambient radiation monitoring programme. These detectors are typically ionization chambers which measure gamma radiation and are configured in a network such that all areas are monitored by at least two detectors. Neutron detectors are also used either in conjunction with or instead of gamma detectors. The detectors sound an alarm if the radiation increases faster than at a preset rate, or if the radiation level exceeds a preset threshold. If two detectors sound an alarm coincidentally, the immediate evacuation alarm goes off automatically. The alarm set point for criticality detectors should be set sufficiently high to avoid false alarms produced by normal movement of material in the facility.

8.9.2. Surface contamination monitoring

Surface contamination monitoring should be conducted periodically at a frequency commensurate with operations and the likelihood of loss of plutonium containment. Such monitoring is conducted to protect the workers directly using handheld radiation detectors and supplemented with swipe surveys. Such surveys are also ideally used for documenting conditions in the workplace. Swipes are typically conducted using a swipe tab and smearing an area of approximately 100 cm^2.

Handheld alpha detectors include gas flow proportional counters, air proportional counters and solid zinc–sulphide ZnS (Ag) scintillation counters.

8.9.3. Airborne activity monitoring

Radiological surveillance of airborne contaminants in the working environment is described in detail in Ref. [18]. Air monitoring is an essential element of the area monitoring programme as it provides sensitive, continuous measurements which verify the integrity of the containment system and early warning to workers if the containment is breached. A comprehensive air monitoring programme uses both continuous air monitors (CAMs) and passive air samplers with filters at the room air exhaust that are installed in the work area to monitor normal work operations. (The filter paper used in a CAM may subsequently be counted like the filters from the room exhaust system.) Additionally, portable CAMs may be used during maintenance operations, or other non-routine conditions where additional monitoring is warranted. These devices make it possible to provide warning or to verify that the facility is being run under satisfactory conditions in terms of airborne radioactivity.

CAMs typically contain a surface barrier silicon semiconductor detector which measures radioactivity collected on a filter paper. Some CAMs use an alpha spectrometer to discriminate between the alpha particles from plutonium and those from naturally occurring radon (^{222}Rn)/thoron and its daughters (which can otherwise cause false alarms). If the level of radioactivity on the filter paper of the passive air sampler exceeds a preset level, the CAM sounds an alarm locally and in the control room. Workers in the area of the alarm should then evacuate the area immediately.

Air sampling is similar to air monitoring except real-time feedback is not provided. That is, the air sampler does not have the capability of assessing the activity on the filter in real-time, so the filters must be sent to a laboratory for analysis. In addition, air sampling provides a low cost means of monitoring the workers and of documenting retrospectively the air quality in working areas.

8.10. INDIVIDUAL MONITORING

The basic requirements for personnel monitoring are described in Ref. [15]. Both the internal and external equivalent doses received by workers must be routinely monitored and recorded, and provisions must be made for assessing doses under off-normal or accident conditions.

8.10.1. External dose monitoring

The routine external dose monitoring programme has historically been conducted using a combination of gamma and neutron radiation dosimeters. Radiation dosimeters must be read on a periodic basis; although monthly exchanges are typical, more or less frequent exchanges may be appropriate depending on the work being conducted and the potential for exposure.

Gamma dosimetry typically uses photographic film or thermoluminescent dosimeters (TLDs). Quartz fibre electroscopes (pen-like QFEs) or digital and audible gamma alarms may supplement film or TLDs when high radiation fields are expected and the operating time must be controlled. Care needs to be exercised when using these devices, however, as they are typically not sensitive to neutron radiation, which may contribute a significant portion of the worker’s dose. Recently, electronic dosimeters have been developed, allowing for real-time external dosimetry and a better knowledge of doses received during a given task [19–24].

Neutron dosimetry may utilize photographic film, track-etch plastic or TLD albedo dosimeters. In any case, the energy of the neutron radiation is important when selecting a dosimeter and assessing the measured neutron dose. Systems capable of providing neutron dose estimates contain a combination of TLDs, activation foils and solid state damage track detectors. Plastic track-etch detectors are also used for fast neutron detection. One example of material currently in use is CR-39 plastic; this material approximates fairly closely the dose response curve in tissue between 0.1 and several MeV.

For workers conducting glove box operations, the dose rate at the hand may be orders of magnitude higher than what is measured at the glove box face. A TLD based finger ring dosimeter is typically used to monitor gamma dose to the hands. However, assessing neutron dose to the hand is far more difficult because of the required size of the dosimeter. Where the neutron dose is significant (relative to the gamma dose), the neutron to gamma ratio can be measured and applied to the gamma dose measured on the finger dosimeter. Another method of assessing neutron doses to the hands involves the use of neutron and gamma wrist dosimeters, which can be used to measure the neutron to gamma ratio; this ratio is then applied to the gamma dose measured by the finger dosimeter.

Special dosimetry is required to measure doses from a criticality accident [25–26]. Nuclear accident dosimetry (NAD) systems are required for areas where a criticality accident is credible, and must be capable of providing information on the neutron spectrum and the dose due to neutrons and gammas at locations around the criticality site.

NAD systems are classified into two categories:

- Area dosimetry systems, capable of furnishing information on neutron spectra and dose at the unit locations;
- Personnel dosimetry systems, containing a minimum number of activation detectors worn as a single badge on the chest or as a belt with the detectors evenly distributed on it.

In addition, the human body itself has proved to be a good personnel dosimetric system for assessing neutron doses by measuring sodium and sulphur activation in the blood and hair, respectively, of exposed individuals.

8.10.2. Internal dose monitoring

A routine comprehensive dose monitoring programme typically uses a combination of urinalysis and lung counting. In the case of a suspected intake, nasal wipes, wound counting and faecal analysis are also employed [27]. The design of a routine bioassay programme is shown in Fig. 7. The methods available are described in Ref. [28].

Routine lung counts are typically conducted on an annual basis. Lung counting is conducted by placing radiation detectors on the outside of the body over the upper lobes of the lungs and measuring the low energy (11–23 keV) X rays released from plutonium inside the lungs. However, the X rays measured are so low in energy that the result varies due to differences in the thickness of the chest wall. This makes accurate measurement of low level plutonium in the lungs very difficult. Recently, a lung monitor with a germanium semiconductor detector has been developed that gives better results and has a limit of sensitivity corresponding to a committed effective dose of about 20 mSv for normal chest wall thicknesses. Although lung counting is not very sensitive, it is a useful component of a routine monitoring programme and a critical component of an emergency response programme as it provides immediate feedback in cases of suspected intakes.

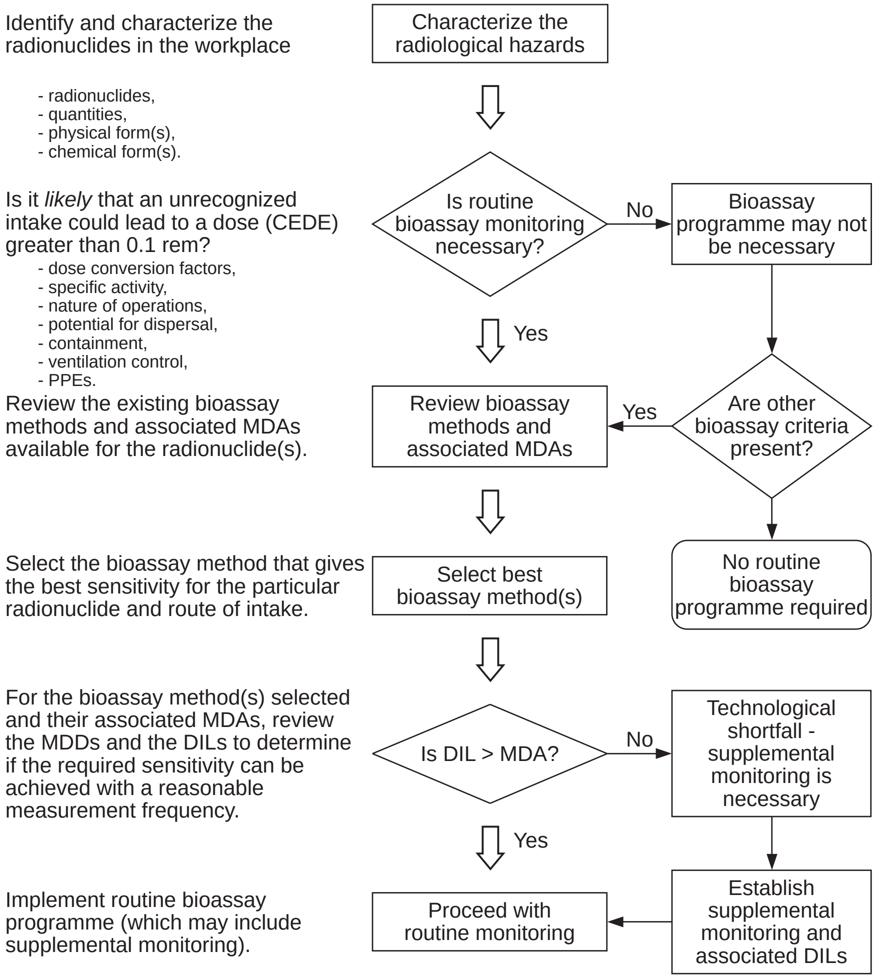
In commercial (LWR) plutonium and some military plutonium, the content of ^{241}Am is sufficiently high to measure the 59 keV gamma rays from it. In this case, internal personal dosimetry can be achieved with reasonable accuracy, but it is necessary to determine the isotopic ratios between plutonium isotopes and ^{241}Am.

Wound counting is conducted in a manner similar to lung counting, except that the radiation detector is placed just above the wound site. Wound counting can provide critical information when determining the amount of radiation in a wound after an accident and the appropriate response.

Personal air samplers are sometimes used as an indicator of intake, but they should not be used to assess personnel dose, as the activity on the filter is often found to be the result of cross contamination, or of a particle which is not of respirable size. The above notwithstanding, personal air samplers are generally considered to be a much more reliable indicator of a worker’s exposure than air samplers at fixed locations as the concentration differential between the workers and the passive air sampler may be two orders of magnitude or more.

Nasal wipes are also frequently used as an indicator of intake, but they should also not be used to assess personnel dose as they are subject to many variables, including whether the person is a nose breather or a mouth breather, how long after the potential intake the wipe is taken and whether or not the person has blown his or her nose. Wipes which are taken more than 15 min after the potential intake are of minimal value, particularly for documenting a zero intake.

For routine monitoring programmes, urine analysis is typically conducted on a semi-annual basis. Although routine faecal sampling can theoretically provide a great deal of sensitivity, difficulties associated with both the collection and analysis of faecal samples usually limit the use of such sampling to follow-up for suspected intake incidents.



Identify and characterize the radionuclides in the workplace

- radionuclides,
- quantities,
- physical form(s),
- chemical form(s).

Is it *likely* that an unrecognized intake could lead to a dose (CEDE) greater than 0.1 rem?

- dose conversion factors,
- specific activity,
- nature of operations,
- potential for dispersal,
- containment,
- ventilation control,
- PPEs.

Review the existing bioassay methods and associated MDAs available for the radionuclide(s).

Select the bioassay method that gives the best sensitivity for the particular radionuclide and route of intake.

For the bioassay method(s) selected and their associated MDAs, review the MDDs and the DILs to determine if the required sensitivity can be achieved with a reasonable measurement frequency.

Implement routine bioassay programme (which may include supplemental monitoring).

FIG. 7. Design of a routine bioassay programme (CEDE: committed effective dose equivalent; DIL: derived investigation level; MDA: minimum detectable activity; MDD: minimum detectable dose; PPE: personal protective equipment; 1 rem = 1.00 × 10^{-2} Sv).

If an intake of plutonium is confirmed either by the routine monitoring programme or following an accident, attempts may be made to reduce the projected dose, as discussed in Section 5.6. Whether or not such attempts are made depends on the magnitude of the dose which may be averted and on input from the worker, the medical staff and health physicists.

8.11. ENVIRONMENTAL MONITORING

Aspects of environmental monitoring are described in detail in Ref. [29]. To ensure that the radioactive release from nuclear sites is within authorized limits, monitoring at the source of discharge is carried out, in addition to which environmental samples are collected and analysed. It is necessary to select sampling items, locations, quantity, season and other conditions based on topography and the geology of the land and of the sea bed around nuclear facilities, atmospheric phenomena and marine phenomena at plutonium effluent disposal sites, the types and quantities of seasonal food to be collected near nuclear facilities, patterns of intake of foods in the area, characteristics of foods produced around nuclear facilities, and data on the behaviour of plutonium in the environment. Sampling locations should be selected where the maximum concentration is estimated to occur.

Pretreatment of samples is needed before analysis for plutonium. The objective is to reduce the volume of the samples. It is important to concentrate the samples without losing the plutonium contained in them.

8.12. OPERATIONAL EMERGENCY PROCEDURES

In this report, the term emergency refers to incidents which do not extend to people outside of the facility. All workers should be adequately briefed on operational radiation emergency procedures, including radiation monitoring, and should take part in exercises so that they will respond correctly in the event of an operational emergency [15].

A systematic, structured approach to the development and implementation of plant specific accident management programmes is described in Ref. [30], emergency preparedness exercises in Ref. [31], medical handling of accidentally exposed individuals in Ref. [32], planning for off-site response to radiation accidents in Ref. [33] and preparedness in Ref. [34]. General principles are described below.

8.12.1. General principles

The objectives in responding to an operational emergency should be to:

- Protect persons from hazard, particularly from inhalation of plutonium;
- Bring the emergency under control;
- Minimize damage to the facility;
- Confine contamination to the smallest possible area;
- Identify persons who were at risk;
- Assess the nature and magnitude of the radioactivity released.

Emergency plans should be prepared beforehand to cover all foreseeable operational contingencies. For example, the following operational emergency conditions can be considered:

- A fire threatening to involve or involving plutonium;
- A spill of materials or solutions containing plutonium;
- Any event that could breach containment;
- Loss of containment owing to:

  (1) Explosion or implosion in a fume hood or glove box,
  (2) Failure of a glove in a glove box,
  (3) Failure of a transfer bag;

- Failure of the ventilation system;
- Power failures that may affect critical equipment and circuits (such as safety related equipment.

The consequences of some operational emergencies have been considered in Sections 4 and 10–12 of Ref. [33]:

- Cuts and wounds contaminated with plutonium,
- Gross skin contamination,
- Inhalation or ingestion of plutonium.

The responsibility for emergency planning and the implementation of those plans should rest with the management, with technical input from specialists in health physics and with medical and fire services. Such planning should cover emergency procedures, including provision of adequate instructions, training and drills (for all staff, but especially for emergency teams) and provision of protective clothing and equipment.

8.12.2. Emergency practical procedures

In zones where there is a potential for fire or explosion, special operating instructions should be provided to identify the hazards and to inform and train the staff. Safety instructions, and procedures to maintain and replace explosion proof equipment must also be provided.

In industrial operations personnel are normally excluded from the glove box regions. When extinguishing plutonium fires in which plutonium is burning or where plutonium may be involved, careful attention should be given to the choice of materials and to the methods used for fire fighting. For localized fires, an eutectic of rare earth salts is commercially available. It should be available inside the box when working with finely divided metals. This mixture fuses at a low temperature and, when placed on the fire, melts and prevents air from reaching the plutonium, thus extinguishing the fire. Other dry powders can be used but may only serve to assist in confining the highly toxic dusts and fumes created by the fire. Materials that should not be used include: water, soda, sodium bicarbonate, foam, carbon tetrachloride, chlorobromo methane and methyl bromide. Carbon dioxide cannot be relied upon to extinguish plutonium fires and its use may significantly increase the dispersal of plutonium bearing dust and fumes.

Dry argon gas or helium can only be used where the compartment involved may be flooded with the gas without causing damage to the containment or filters.

Other incidents of a minor nature occur more frequently and certain practical measures are necessary to:

- Prevent the internal contamination of the individual,
- Restrict the spread of the contamination in the working environment,
- Control the source and reduce the consequences of an incident.

8.13. EXAMPLES OF INCIDENTS AND PRACTICAL RESPONSES

8.13.1. Failure of a glove or transfer bag

If a serious failure of a glove occurs while working at a glove box, *do not* withdraw your hand. Place a respirator, if available, over the mouth and nose with a free hand, or shout for respiratory protection to be provided and then hold your breath if possible until the respirator is provided. With respiratory protection on, withdraw your hand, leaving the glove inside the box. The hand should be monitored with instruments and wrapped in a plastic bag before the worker is sent for decontamination. The damaged glove should be replaced as soon as possible. In the case of a badly torn transfer bag or a glove which is completely torn away from the glove port, use an emergency plug if available.

8.13.2. Excessive pressure in a glove box

Respiratory masks (or other devices to prevent inhalation) should be available and used if the operating pressure of the glove box becomes less negative than normal, which is about 25 mm WG of negative pressure (see Fig. 6). If possible, connect the box to another one, for instance via the transfer tunnel, open the exhaust damper fully and adjust the inlet damper until the normal negative pressure is achieved.

8.13.3. Excessive negative pressure in a glove box

If possible, connect the box to another one, shut the exhaust damper and adjust the inlet damper until the normal negative pressure level is achieved (connecting to a much smaller glove box may have little effect). If gloves or transfer bags fail as a result of such an incident, respiratory protection should be made available and used.

8.13.4. Contamination of protective clothing

The person wearing the contaminated clothing should put on respiratory protection and possibly have his/her suit sprayed to reduce the resuspension of dust which could arise from the removal of the clothing. Contaminated clothing can be placed in a plastic bag to prevent the spread of contamination; after removal of the contaminated clothing, the person should be surveyed for skin contamination.

8.13.5. Local skin contamination

Wash with tepid water and non-corrosive soap and, for hands, use a soft nail brush. Rinse and dry thoroughly and recheck for contamination (see Section 5.4). Refer to the medical services if contamination persists.

8.13.6. Contaminated wounds (see Sections 5.6 and 8.11)

For serious wounds, conventional first aid treatment will be necessary, and the person should then be referred to the medical services. For minor wounds thorough irrigation should be carried out and the person referred to the medical services.

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9. SUMMARY

9.1. SAFE HANDLING OF PLUTONIUM

Plutonium is a highly radiotoxic element which can be utilized to produce significant benefits if handled and stored safely. This report provides an overview of the types and quantities of plutonium present in different countries, the potential hazards associated with the material and the safety considerations associated with the handling and storage of significant quantities of plutonium.

The most important factors to keep in mind when evaluating systems or operations utilizing plutonium are to:

- Avoid criticality,
- Avoid fire and explosion,
- Maintain containment to avoid contamination,
- Avoid internal exposure to plutonium,
- Ensure safeguards,
- Provide adequate physical protection,
- Keep external exposures at ALARA levels.

The primary approach to plutonium safety involves planning, personnel practices and engineered controls, such as the use of glove boxes, remote handling systems, geometrically safe containers, HEPA filtration and dynamic ventilation systems. Further safety considerations include administrative standards and controls such as mass limitations, training, procedures, postings, personnel and area radiation monitoring, and emergency response. The combination of all these elements provides the basis for a comprehensive safety programme.

9.2. STORAGE OF PLUTONIUM

The storage of plutonium entails ongoing attention, as the heat and gases generated during storage can be significant. Safe storage can be achieved if:

- Mass and geometry controls are observed.
- Cooling is provided for storage vaults.
- For long term storage (longer than one year), plutonium is stored as the oxide (PuO_2), metal, stable alloy, or stable compound (not yet identified) in a sealed container; it is necessary to use the proper atmosphere, such as a vacuum or dry inert atmosphere.
- Storage containers are kept free of organics.

- Storage containers are kept free of moisture.
- Ongoing monitoring, surveillance and maintenance are conducted.
- Emergency procedures are developed and practiced.

A significant level of expertise has been gained over the last 50 years on the safe handling and storage of plutonium — particularly with high ^{238}Pu and ^{241}Am content. This expertise allows large scale operations to be conducted safely, relative to workers, the general public and the environment.

Annex I

EXAMPLES OF PLUTONIUM PLANT DESIGN

I–1. INTRODUCTION

Production of civil plutonium is now in its fourth decade and equipment and systems are at an advanced state in terms of:

- Flowsheets,
- Containment of plutonium,
- Minimizing effluents,
- Quality control,
- Minimizing the risk of fire and explosion,
- Minimizing the dose to operators (remote operation),
- Minimizing the dose to the public,
- Radiological safety,
- Criticality safety,
- Industrial/conventional safety,
- Safeguards,
- Physical protection.

Section 7 outlined general considerations in the design of a plant and Section 8 outlined operational safety in the plant. As discussed in Section 6, all such undertakings are in strict compliance with the conditions of the ‘License issued by the Competent Authority’. Enforcement of the license, and compliance with documentation issued by the company to support safe operations, is carried out by the Competent Authority which routinely inspects all nuclear licensed sites.

The examples in this annex illustrate how some of these considerations are incorporated into actual plants and operations for the cycle from conversion, packaging and storage, through to MOX fuel production.

I–2. MODERN GLOVE BOX DESIGN

Glove box design will be dependent on the specific processes undertaken in that box, but general principles apply to all such boxes. Modern glove boxes are usually made of welded, heavy gauge, micropolished stainless steel, which provides some gamma shielding, a high degree of leak resistance and easy cleaning, making very

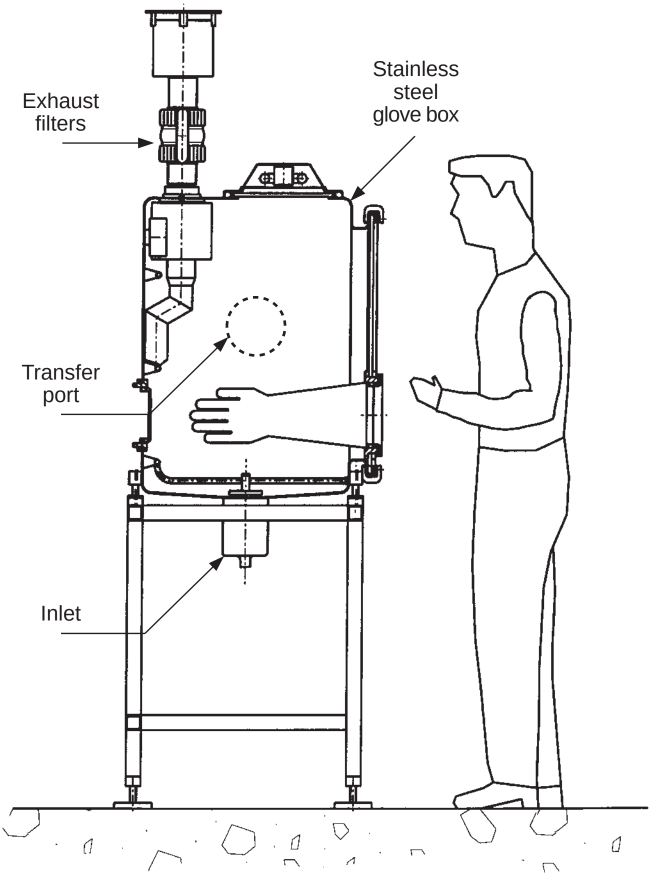

FIG. I–1. Side view of a typical glove box.

low fire loading possible (see Fig. I–1). Windows are usually small in area relative to the box face size and incorporate laminated lead glass. Plastics with and without lead are also used in some windows. Replaceable internal screens for the windows are incorporated if internal impact sources are present.

The gloves fitted to the box are usually ‘hypalon’. These come in a range of thicknesses, lengths and with lead loading if required. Mountings on the glove box can incorporate spigots or push through cartridge units to facilitate glove changing.

Additional neutron shielding of bulk sources may be required — steel clad polythene is routinely used. For glove boxes handling plutonium from high burnup spent fuel or with, for example, plutonium fluorides present, additional external

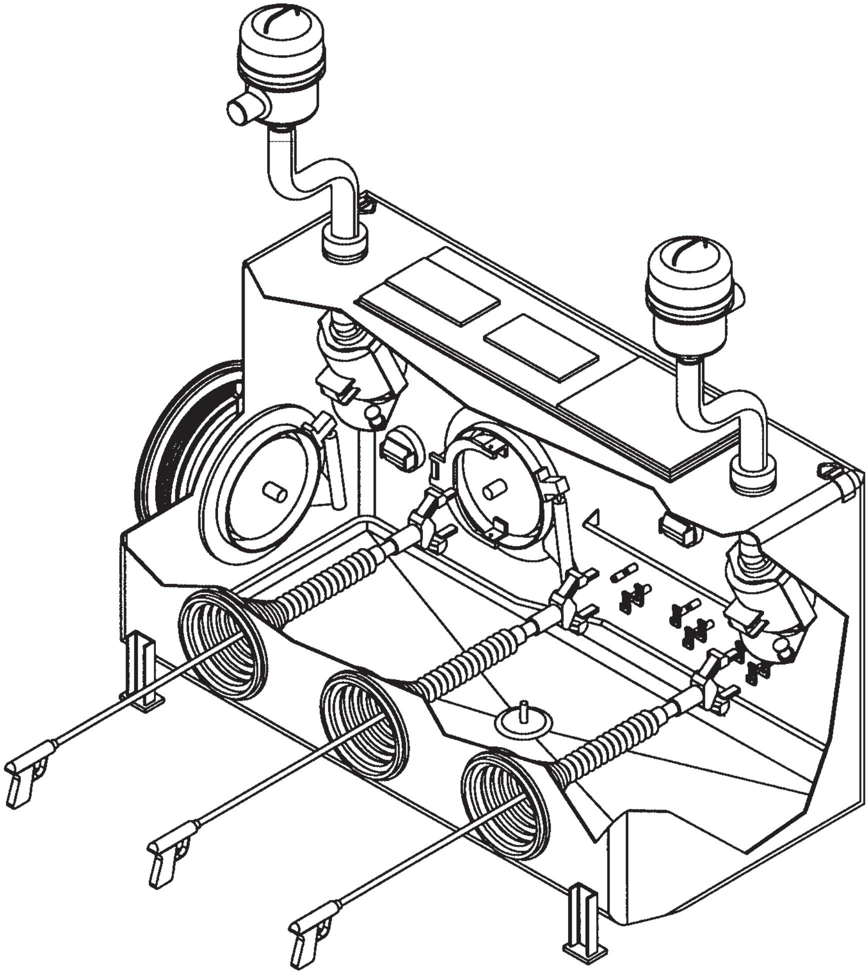

FIG. I–2. View of a typical glove box with manipulators for an analytical laboratory.

neutron shielding may be necessary to limit dose rates to operators. Clad polythene or fire resistant Jabroc (a compressed laminated beechwood) can be used as neutron shielding.

The shielding of glove ports when not in use is also incorporated as a way of minimizing the dose to operators. Such shields may incorporate neutron shielding, e.g. metal clad polythene or simple gamma shields such as steel. In industrial operations workers are normally excluded from glove boxes during routine operations. Where workers are required to work at glove boxes, gloves can be replaced by master–slave manipulators to reduce the exposure (see Fig. I–2).

Ergonomic considerations are incorporated into the positioning of the glove ports, windows and items within the boxes at an early stage of the design. As well as ensuring that spacing matches the average/standard operator, task analysis ensures that the full range of operations necessary can be accommodated. Maintenance operations, in particular, require good visibility and accurate placing of glove ports. Confirmation of the correct placing is achieved during plant construction and commissioning. The glove box size should be limited to the minimum necessary.

To transfer items to and from the glove box it is possible to use either traditional welded bag methods or bagless transfer units that maintain glove box integrity during transfer into a container fitted with a double lid. A range of port sizes may be fitted, which require shielding and protection when not in use. Transferring items in (termed ‘posting in’ in some countries) can also use traditional bag methods, simple interlocked airlocks and sphincter methods.

Glove boxes can be illuminated by the use of re-entrant tubes containing fluorescent lights. These facilitate the replacement of failed lights or tubes without breaching the containment. Internal lighting by this method is much more flexible than lights that are external to the box windows.

Electrical services passing into the glove box make use of permanent through-wall connectors rather than glanded penetrations. Gaseous and liquid services also have permanent penetrations; self-sealing couplings are often provided, with filtration and automatic isolation valves fitted to these supplies to prevent back diffusion of glove box activity or box pressurization. Motor drives are usually external to glove boxes with simple push-through wall cartridge units or sealed magnetic couplings that can be replaced without breach of containment.

For glove boxes containing liquids, sloping floors with sumps and drains to criticality safe storage vessels should be installed. The sumps are fitted with alarm devices to warn of arisings and to isolate supplies as necessary.

Local filtration of glove box ventilation to protect ductwork from contamination is provided by HEPA filters that can be replaced without breach of containment. During bleeding operations, systems are similarly fitted with HEPA units to prevent back diffusion. Fluidic devices, which have no moving parts, can be used to control glove box depression while providing a rapid increase to the air flows to compensate for a failed glove.

Operational equipment within the box is provided with all normal conventional safety features, but in addition requires close examination of certain features. These include:

— Corrosion resistance,
— Absence of sharp points, edges or pinch points,
— Engineering-out features requiring detailed maintenance using tools such as screwdrivers and knives,

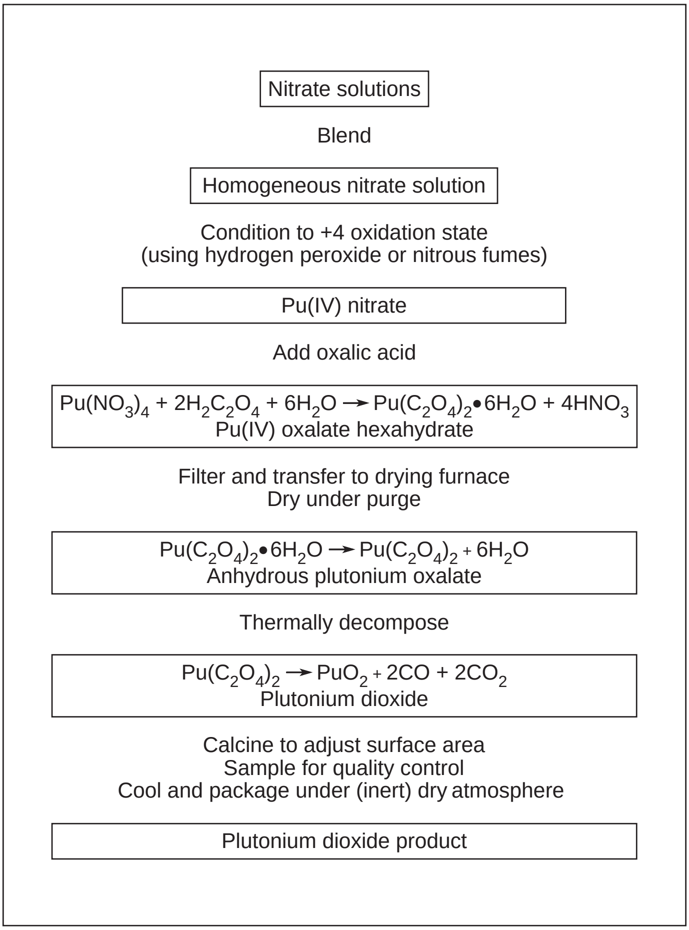

FIG. I–3. Scheme for conversion of plutonium nitrate solution to plutonium dioxide product.

- Lubrication free units,
- Use of low voltage equipment when possible,
- Spark resistant equipment,
- Unit replacement through transfer ports,
- Guarding of powered items,
- Avoidance of stored energy items,
- Absence of cavities capable of holding or retaining plutonium.

I–3. CONVERSION (FINISHING)

Plutonium production at the two major reprocessing facilities in the United Kingdom and France is based on the precipitation of plutonium oxalate from plutonium nitrate solution after chemical separation from fission products and uranium. The examples given are for the United Kingdom, but differences are pointed out. This process has also been used historically for the production of military material, except that the oxide may be subsequently fluorinated before reduction to metal. A typical scheme for conversion of plutonium nitrate solution to plutonium dioxide product for storage and subsequent fuel fabrication is shown in Fig. I–3.

Pipes for the transfer of solutions are constructed from seamless, nitric acid-resistant, stainless steel to minimize the risk of breach of containment due to corrosion. The integrity of the piping is extensively tested during construction and commissioning.

Glove boxes are constructed from seamless stainless steel to minimize the uptake of dust and to facilitate cleaning. Process vessels within glove boxes act as an initial level of containment to minimize activity which can find its way onto the inside faces of the glove boxes. The process vessels themselves are shielded, where possible, rather than applying heavy shielding to the whole glove box or cell. In this way, the amount of shielding required on the faces of the box can be minimized, which improves visibility and manœuvrability for maintenance work.

Operations are carried out remotely, with manual intervention only required in the event of breakdown. Prior to any maintenance work the vessels and glove boxes/cells are cleaned out to minimize the dose to operators.

Blending of solutions is carried out (within geometrically safe vessels) to ensure a product with uniform isotopic composition. The cells which house these vessels contain a sump which is designed to accommodate any leak without liquid accumulating to form a critical geometry.

The plutonium nitrate is conditioned to the Pu(IV) oxidation state to optimize oxalate precipitation. In the United Kingdom, this is done by feeding the nitrate solution into a second series of vessels where it is mixed with hydrogen peroxide solution. Several vessels are used in parallel, rather than one large vessel, to keep the volume to one which is criticality safe. In France, the valency adjustment is achieved upstream by the use of nitrous fumes after separation.

The plutonium nitrate is fed to another set of geometrically safe vessels where a slight excess of oxalic acid solution is added to cause precipitation of plutonium di-oxalate hexa-hydrate. The crystals are allowed to age for about 15 min to increase the average particle size to aid filtration (the ageing time will depend on the temperature, concentration, acidity, etc.).

The precipitate overflows and passes to a rotating vacuum filter. The volume of material on the filter is restricted by a cover plate as well as by the operation of a

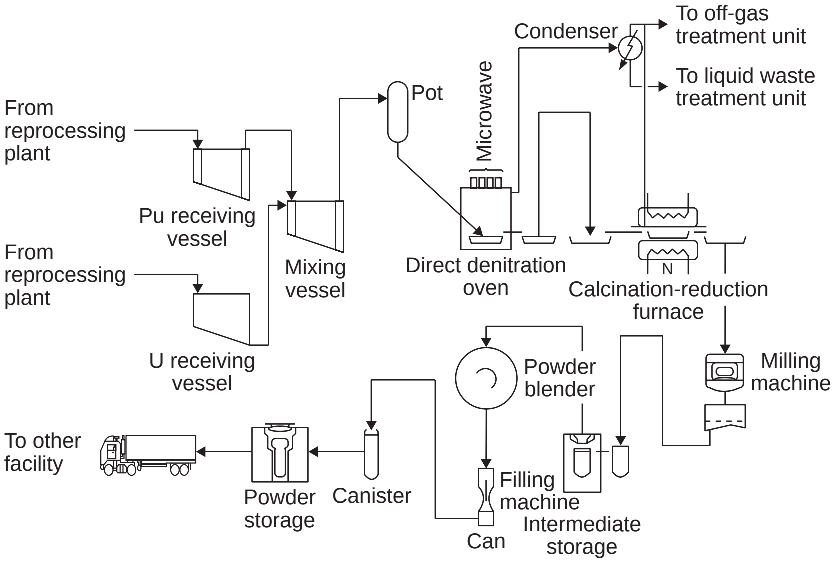

FIG. I–4. Schematic diagram of the co-conversion process used in Japan to fabricate MOX fuel.

scraper bar. This bar continuously removes the precipitate from the rotating filter and deposits it down a tube into a drying furnace.

The filtrate contains residual plutonium. This is concentrated by evaporation and then refluxed with concentrated nitric acid to destroy oxalic acid. The plutonium bearing solution is then returned to the chemical separation area to minimize effluent arisings.

The oxalate is moved through a temperature profile in the furnace by an internal Archimedes screw. The oxalate is dried, dehydrated and then thermally decomposed as the temperature increases along the length of the furnace. A countercurrent air flow passes over the drying oxalate to remove moisture and decomposition products. The purge gas is extracted and filtered in a separate line and does not enter the glove box suite, thereby avoiding the plate-out of dust. The furnace is built to withstand a deflagration from the reaction of carbon monoxide from the decomposition of oxalate. The plutonium dioxide is calcined to reduce the surface area to around 10 m^2/g in the final stage of the furnace or in a separate furnace.

In Japan, a ‘co-conversion process’ has been developed to enhance the non-proliferation properties of the MOX fuel cycle. In this process the plutonium nitrate from the reprocessing facility is mixed with uranium nitrate and then heated by microwave to produce the mixture of oxides (see Fig. I–4). This process requires no unique safe handling procedures.

I–4. PACKAGING

Plutonium dioxide can readily absorb gases and moisture onto its surface. In the United Kingdom, it is cooled and packaged under a dry argon atmosphere. Samples of oxide are taken prior to packaging to demonstrate that moisture levels are below specified levels for storage. These moisture levels are limited both to avoid criticality and to avoid pressurization of packages from chemical or radiolytic degradation. Similarly, the amount of carbon, either from residual oxalate or organics, is restricted but this is not a problem when calcination is carried out under air.

In the British Nuclear Fuels Limited (BNFL) plant for low burnup Magnox derived plutonium, the plutonium dioxide is first delivered and weighed into a 3 L, screw top aluminium can which acts as the first level of containment. The French plants use a first stage stainless steel can with a capacity of about half of this.

For low burnup fuel, such as Magnox, transfer from the contaminated to the uncontaminated areas is effected by bagging the primary can out of the glove box suite in a polyethylene secondary containment using conventional bagging and welding techniques. The aluminium can in the polyethylene intermediate is then placed inside a steel can and a lid is resistance welded in place.

The integrity of the polyethylene intermediate is destroyed either during welding or by radiolytic and thermal degradation during storage. Decomposition of the polyethylene has not led to any pressurization of the container or degradation of product quality.

In the past, PVC has been used as the outer secondary containment, but when this degrades it has caused chloride corrosion of the outer stainless steel can. Oxides stored with PVC had to be recalcined to remove absorbed chlorine and repackaged in chloride free systems. For plutonium derived from high burnup fuel, the oxide temperatures can exceed 100°C at the can wall, with up to 25 W/kg of decay heat being generated. These cans cannot be manually handled at all and cannot be allowed to come into contact with organic materials. For these oxides, can filling is as before, but into a stainless steel inner can. Here the transfer from contaminated to uncontaminated zones is achieved by: sliding a clean stainless steel intermediate can into the neck of a sphincter seal between the boxes; inserting the inner can into the intermediate can, inserting a bung; welding the bung into the intermediate can by rotating it in the beam of a laser; refocusing the laser; and rotating the can in the beam to effect a cut. The intermediate can, which has remained clean on the outside, is then transferred into the clean box, while the remains of the bung seal the sphincter against release of contamination. A new intermediate is inserted to displace the contaminated bung into the contaminated box. The beam of the laser passes through an optical window so that the equipment is remote and easily maintainable. The intermediate can is then placed inside another stainless steel can and a lid is resistance welded into place. The three levels of close fitting stainless steel afford good heat dissipation.

Proof testing of package designs involves destructive testing by pressurization of empty containers under normal and fault conditions, i.e. after drop tests, and at normal and elevated temperatures. The packages withstand greater than 20 atm of internal pressure and even higher external pressure. The final product is leak tested to demonstrate compliance with international transport requirements.

I–5. STORAGE

A key issue in the storage of separated plutonium is the extent to which large stores of plutonium constitute a proliferation risk. Accordingly, storage facilities are designed to ensure safe, secure storage, while satisfying safeguards. These facilities have been constructed with massive, secure containment, robust packages and high densities to economically control heat generation and radiation. Such storage facilities are typically submitted for safeguards verification and multiple systems of containment and surveillance with redundancy to form an effective safeguards system.

Plutonium dioxide is stored in small leaktight packages, as described, and these are placed using remote mechanical means in racks or channels in a rigid matrix within massive concrete cells. Such an arrangement maintains segregation to avoid criticality incidents and prevents physical damage to the packages during handling from external hazards such as aircraft crashes, extreme weather and seismic events. Siting of the store is chosen to minimize external hazards. In addition, the cells provide significant gamma and neutron shielding and security barriers; the materials have a minimum fire loading. Removal of the decay heat relies on the good thermal conductivity of the package and forced ventilation through the racks or channels. High levels of reliability of the ventilation systems are achieved by reinforced back-up power supplies and systems with high levels of redundancy in the control and extraction systems.

The design capacity of the system is for a full store at maximum controlled decay heat loading. Demonstration of the ability of the packages and store to survive ventilation failure is part of the licensing process and may be different depending on the source of the plutonium stored. For low burnup derived Pu, remedial engineering work and partial restoration may be possible; with high burnup derived Pu, a non-interventionist approach may be necessary, relying on natural convection for cooling (i.e. a passive safety principle). In both cases the package performance will depend on the analysis and control of the product against a specification and the quality control of the materials and sealing of the package.

Containment is ensured by the design of the packaging and by testing its performance under normal and fault conditions. Monitoring and filtration of the store ventilation extract is undertaken to ensure that there is no breach of the packages.

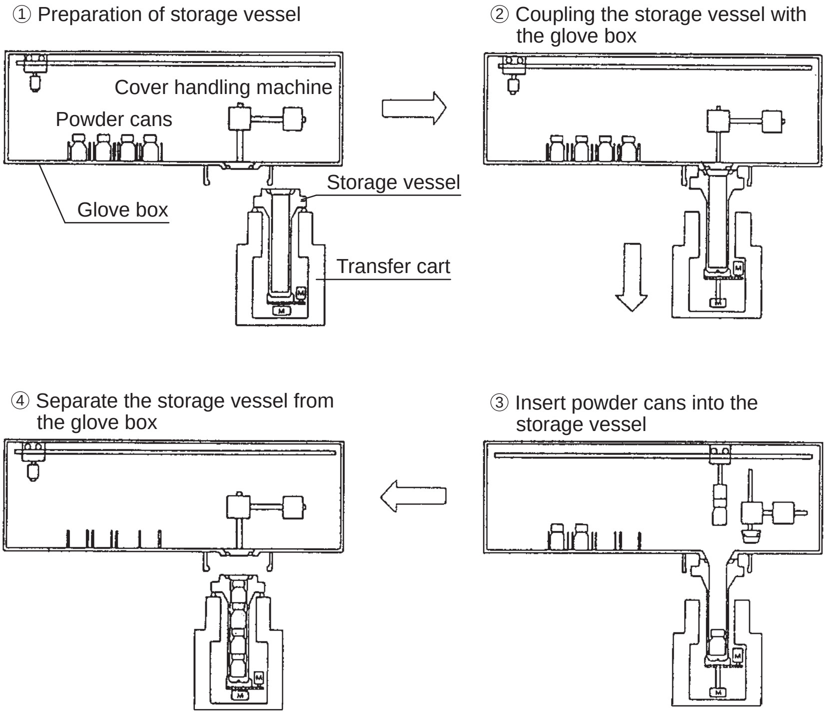

FIG. I–5. Schematic of the double cover plutonium storage system.

Simple visual and dimensional inspections of the packages in their storage position may be carried out using remote devices.

Storage of plutonium bearing residue materials prior to processing may require additional characterization and stabilization of the materials to eliminate the risk of package pressurization or corrosion. Heat treatment in an oxidizing atmosphere or air to about 500°C and cooling in a dry atmosphere will remove moisture and destroy organic contaminants sufficient for interim storage.

Plutonium from weapons sources may be stored in metallic form, so a somewhat different set of packaging requirements is recommended based on experience in the USA [I–1]. To minimize the risk of pyrophoric ignition, metal pieces should have a minimum thickness of 0.5 mm and excess oxide should be brushed from the surface. Large pieces of metallic plutonium, while less likely to ignite, will on oxidation exhibit a significant expansion capable of bursting the packaging or container. Packaging is completed by placing the metal in an approved stainless steel container, purging with dry inert gas and sealing the container by welding and certifying the integrity of the seal by leak testing. An additional margin of safety is achieved by a second sealed containment.

Storage facilities incorporate design features to assist the verification of the contents by the operators, the IAEA and Euratom. These include, in addition to security devices, the provision of secure seals for the storage channels and stores, continuous monitoring, surveillance and recording equipment, weighing and non-destructive analysis equipment to assess storage container contents and the ability to inspect containers within the channels. In the THORP store and the Magnox store under construction, remote identification of containers within the channels is also possible.

The physical security of plutonium facilities is ensured by compliance, at a minimum, with international standards [I–2] and conventions [I–3]. Meeting these standards, plus additional national government requirements, is achieved through the methods and material of construction, the installation and operation of appropriate surveillance equipment and the strict control and limiting of access through a secure perimeter to achieve protection in depth.

In Japan’s Tokai MOX fuel plant, MOX powder is put in aluminium cans, and these cans are then inserted into stainless steel storage vessels with a double cover. The entire operation is carried out remotely, as shown in Fig. I–5.

I–6. SAFETY PRINCIPLES APPLIED TO MOX FABRICATION

Mixed oxide fuel for thermal reactors uses fissile plutonium in place of some of the ^{235}U. MOX fuel fabrication is performed on a commercial scale in Belgium [I–4], France [I–5], Japan and the United Kingdom [I–6]. The typical steps of a MOX fuel fabrication process are shown in Fig. I–6. The most commonly used first process step consists of blending plutonium oxide and uranium oxide. Other processes use co-conversion of uranium and plutonium nitrates to mixed oxides in the first step (see Figs I–4 and I–5). In both cases, the following steps (from pelletization to fabrication of the fuel assembly) are very similar to those used to produce uranium oxide based fuel for LWRs, so handling practices are similar.

This section gives concrete examples of the application of safety design principles to commercial scale MOX fabrication facilities. The text deals primarily with the MELOX facility in Marcoule, France, though significant differences in other plants are indicated.

In 1996, the nominal yearly capacity of the plant was about 120 t of mixed uranium and plutonium oxide fuel for LWR reactors. The plutonium handled in the MELOX facility must not contain more than 3% ^{241}Am by weight (to control the gamma dose) and the ^{240}Pu content must be at least 17% (to help control criticality). The ^{241}Am and ^{240}Pu are controlled in other plants for the same reasons but the

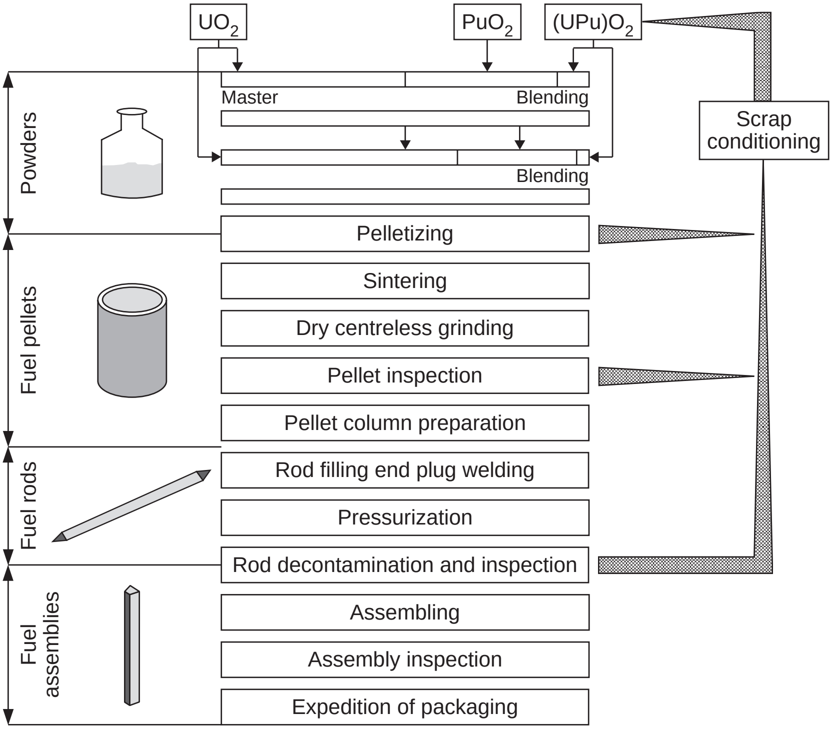

FIG. I–6. Typical MOX fabrication process used in France and the United Kingdom.

amounts will be different. For example, the ^{240}Pu in weapons plutonium will be about 7%. The plant consists of the following:

- A fuel assembly fabrication building for the receipt of raw materials, powder mixing, fabrication of fuel rods and assemblies, and inspection and acceptance testing;
- A building devoted to waste treatment, including an incinerator for alpha contaminated technological waste. After treatment, the waste is conditioned and sent either to the La Hague Cogéma plant for plutonium recovery, or to a final storage centre.

The fabrication process adopted is derived from those that have been in use in the Belgonucléaire (Dessel, Belgium) and Cogéma (ATPu) plants. The MOX pellets are produced by blending uranium and plutonium oxide powders, the mix initially having a high plutonium content of between 25 and 30 wt% (master mix), which is then diluted with uranium oxide to obtain the desired proportion (between 3 and 11 wt% Pu). BNFL's short binderless route achieves the desired enrichment in one step using a high throughput attrition milling stage to obtain the desired PuO_2/UO_2 proportions.

I–6.1. Contamination

Static containment to prevent dispersion of radioactive contamination is provided by three physical barriers between the material and the environment: for powder, the first is constituted by the glove boxes or cells, the second by the room walls and the third by the building. The dynamic containment provided by the ventilation system creates a cascade of pressure differentials from the parts of the installation with the highest risk of dispersion towards those with the least. Several ventilation systems are provided (see Figs 4–6 and Section 7.3 in the main text):

- The highest is the process equipment,
- A high negative pressure system for glove boxes (P = –25 mm WG),
- An intermediate negative pressure system (P = –8 mm WG) for the ventilation of rooms containing glove boxes,
- A slightly negative pressure system for the other rooms.

The air extracted from the glove boxes passes through three filtration stages before release. In addition, arrangements are made to ensure that the ventilation will not fail, essentially by adequate redundancy of the electrical power supplies, including emergency generators. The overall objective of these measures is to guarantee that contamination of the premises is zero during normal operation. A high sensitive air monitoring network and associated alarms ensure early detection of an abnormal situation (see Section 8.10).

Sintering reduces the risk of contamination when pellets are inserted in fuel rods; the zircalloy tube constitutes the primary containment barrier.

I–6.2. Radiation exposure

Very conservative measures have been taken into account. During the plant design, the objective was to limit the average dose of operating personnel to less than 10% of the permissible dose (5 mSv/a instead of 50 mSv/a). This is achieved by:

- A high degree of automation with remote control rooms to keep operators away from sources of radiation;
- Use of local shielding on the glove boxes, or preferably directly around the equipment;

- Limitation, from the design stage, of the number and duration of operations to be carried out on process equipment (accessibility and visibility constraints in the glove boxes, standardization of equipment, design of mechanical equipment in the form of assemblies of replaceable modules, providing special means for carrying out work, etc.).

I–6.3. Criticality

The study of criticality hazard is based on the fissile material content of the blended oxide powders mentioned earlier. Criticality control methods are as follows:

- Mass and moderation control for the part where powders and sintered products are used. Three kinds of powder are considered: pure PuO_2, mixed UO_2/PuO_2 with 30 wt% of plutonium (master mix), or with 12.5 wt% of plutonium (upper limit for thermal fuel),
- Geometrical arrangement (mass and spacing) for the storage of powder, pellets and fuel rods,
- Geometrical arrangement (mass and spacing) and moderation control for the storage of fuel assemblies,
- Mass for the incinerator.

Thus, one or more limitations are imposed per workstation or item of process equipment. In addition, the fuel assembly fabrication building is fitted with a criticality incident detection network and alarm systems.

I–6.4. Decay heat

Decay heat hazard is taken into account for all storage of nuclear material (mainly because of the ^{238}Pu content). Cooling is provided by air flow. The ventilation system is also designed to help remove the decay heat of nuclear material in glove boxes.

I–6.5. Radiolysis

The radiolysis hazard is very important in aqueous plutonium solutions, but it may also occur in the presence of organic compounds. The plant does not handle plutonium solutions. This hazard is, however, to be considered for fabrication rejects that are likely to contain organic additives: measures are taken to eliminate these additives.

I–6.6. Fire and explosion hazard

Fire and explosion must be considered to be a major hazard for a plutonium facility as there is a potential to breach the containment and cause the release of radioactive material to the environment. Control of the fire hazard depends on the suitability of the arrangements taken to minimize the risk, to detect the outbreak of a fire and to mitigate the consequences. Thus, an effort is first made to reduce the fire load densities of the facilities and to use fire retardant materials, e.g. for electric cables. Details of fire protection are plant specific, but the general philosophies are similar.

The facilities are monitored by an automatic fire detection system triggering an alarm in a control room to alert an emergency team. In view of the restrictions imposed by the criticality hazard, action can be taken using low hydrogen content extinguishing powders, carbon dioxide or halon (which is a trade name for a series of halogenated compounds). It must also be pointed out that the presence of an inert gas atmosphere (nitrogen) in the glove boxes reduces the risk of fire.

In rooms containing significant amounts of fissile material in glove boxes or with a high fire load density, a fixed CO_2 extinguishing system needs to be installed. This is the case for rooms intended for milling, mixing, granulation, pressing, sintering, grinding and sorting. This system includes two storerooms containing CO_2, located inside the building. Lastly, as a stand-by, an additional extinguishing system can supply CO_2 from outside the building. This system is connected above the nozzles of the fixed system.

Allowance for the fire and explosion hazard in the design of the installation determines the way the facility rooms are arranged, particularly those where powders are handled. The rooms are arranged to form fire and containment zones. Any fire, with the associated radioactive materials, is thus contained within the corresponding fire containment zone. In the MELOX plant, these zones are able to contain an internal fire for at least 2 h. This time is compatible with the present fire load density. To maintain extraction of the air from the zone affected by the fire for as long as possible, the plant extraction network is arranged to ensure dilution of the hot air extracted so as to protect the last filtration level before release to the environment (the dilution factor is between 8 and 10 according to the system). In addition, the extraction ducts are designed to withstand fire. A fire jeopardizing the integrity of the containment zone is taken as a design basis accident for the plant to demonstrate the validity of the choices made.

The prevention of explosions deals only with hydrogen, as there is no organic solvent in this facility. Hydrogen (5%) in argon, mixed outside, is used in the sintering furnaces. Hydrogen gas detectors are installed in the facilities where the hazard is present and the ventilation conditions allow dilution in the event of leakage. This risk of external explosion hazard results from river transport on the Rhône River. The maximum credible accident is the explosion of a barge for the transport of hydrocarbons with inadequately ventilated empty tanks. The resulting pressure wave of around 45 Pa is allowed for in the design basis of the civil engineering structures.

I–6.7. Seismic hazards

The SSE adopted for the site is of an intensity of VIII–IX on the MSK scale, with two resonance spectra:

- One type-1 spectrum (shallow earthquake) set at 0.3g,
- One type-2 spectrum (deep earthquake) set at 0.2g.

These spectra are used as the design basis for those parts of the installation that are important for safety (containment and prevention of criticality hazard). The principles selected for the design basis of the equipment seek to:

- Ensure operation of the intermediate pressure ventilation system after an earthquake,
- Ensure the subcriticality conditions for the equipment containing Pu,
- Ensure emergency power supply.

I–6.8. Aircraft crash hazard

The design objective is that the global probability of an airplane crash causing unacceptable damage to the environment does not exceed 10^{-6} per year.

I–6.9. Minimization of waste

As in any industry, nuclear fuel fabrication generates process scraps and waste. The objective is to recycle most of the scraps in the process line and recover as much plutonium as possible from the waste to reduce the volume and toxicity of the waste and compact it for final conditioning.

An on-site auxiliary building is devoted to waste and scrap conditioning, including a large incinerator with a capacity of 20 kg/h for the alpha contaminated burnable waste. When the scraps are not directly recycled, their ashes are chemically treated in the La Hague reprocessing plant.

Intensive developments have been made on the incinerator, especially to test filter efficiency: these filters must retain more than 99% of dust from the exhausted effluent. A half-scale inactive pilot plant has been constructed for that purpose and tests performed for more than one year.

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Annex II

RADIOLOGICAL SAFETY DATA FOR PLUTONIUM HANDLING

II–1. INTRODUCTION

Important sources of external radiation exposure in handling plutonium are listed below:

- Gamma photons and spontaneous neutron emissions from plutonium isotopes and gamma photons from ^{241}Am. The fuel burnup determines the isotopic composition of plutonium and radiation yield; the amount of ^{241}Pu and the time since reprocessing determines the subsequent amount of ^{241}Am.
- Neutron emissions from (α, n) reactions with oxygen, carbon, fluorine in impurities of oxides, carbides and fluorides, respectively.
- Hard gamma photons are emitted primarily by ^{208}Tl and ^{212}Bi, which result from multiple recycled plutonium containing ^{236}Pu. The ^{236}Pu decays to ^{232}U and leads to exposure problems associated with the ^{232}U decay chain.
- Gamma photons emitted by the fission product ^{106}Ru. The usual specification for all fission products is that the gamma activity should be less than 3.7×10^4 Bq/g Pu (1 µCi/g Pu).

II–2. SURFACE GAMMA DOSE RATE OF PuO_2 AND MOX [II–1]

The effective surface dose rate of plutonium is defined as the dose rate through relatively thin layers of protective containment materials, such as plastic, which are used to contain the toxic plutonium compounds. The surface gamma dose rate equation for plutonium metal or oxide is given below:

$$D_s\ (\text{mGy/h}) = (1710 \times f_{238}) + (5.1 \times f_{239}) + (24.0 \times f_{240}) + (87.0 \times f_{241}) + (1.5 \times f_{242}) + (0.74 \times f_{241})\, t$$

where

D_s is the surface dose rate of plutonium metal or PuO_2 (mGy/h),
f_i is the weight fraction of the ith isotope of plutonium,
t is the time since chemical separation (d).

TABLE II–1. GAMMA ATTENUATION FACTORS OF VARIOUS MATERIALS FOR PuO_2 SOURCES [II–1]

Shield materials	Thickness (cm)	Attenuation Factors[a] Source A[b]	Source B[c]
Stainless steel	0.013	0.48	0.93
	0.038	0.38	0.77
	0.075	0.29	0.58
	0.147	0.18	0.38
	0.331	0.08	0.18
	0.475	0.06	0.13
	0.635	0.05	0.11
	1.351	0.02	0.05
	2.169	0.011	0.03
	2.578	0.007	0.02
Lead	0.005	0.58	
	0.015	0.04	0.22
	0.335	0.014	0.051
	0.503	0.008	0.030
	0.665	0.005	0.021
	1.341	0.0013	0.008
	1.676	0.001	0.005
Lead glass	0.635	0.023	0.071
(4.91 g/cm^3)	1.270	0.009	0.040
	1.905	0.005	0.017
	2.565	0.003	0.010
Neoprene glove	1 layer	0.84	0.90
(0.038 cm nominal thickness)	2 layers	0.72	0.89
	3 layers	0.62	0.88
Lead loaded neoprene	1 layer	0.26	0.51
(0.076 cm nominal thickness)	2 layers	0.14	0.30
Acrylic plastic	0.150	0.935	0.90
	0.455	0.80	0.89
	0.635	0.75	0.88
	0.953	0.66	0.81
	1.354	0.60	0.80
	1.981	0.52	0.79
	2.718	0.48	0.75

[a] Attenuation factor = shielded dose rate/unshielded dose rate.

[b] PuO_2 in source A from Yankee Rowe (PWR); this source produced 45 mGy/h unshielded.

[c] PuO_2 in source B from Dresden (BWR); this source produced 15 mGy/h unshielded.

TABLE II–2. NEUTRON YIELDS FROM Pu ISOTOPES[a] AND COMPOUNDS [II–1 to II–3]

Isotope/ compound	Spontaneous fission half-life (a)	υ (neutrons per fission)	Spontaneous fission yield (n/s per g)	(α, *n*) reaction yield (n/s per g)
^{236}Pu	3.5×10^{9}	2.3	3.7×10^{4}	
^{238}Pu	4.9×10^{10}	2.33	2.62×10^{3}	
^{239}Pu	5.5×10^{15}	3.0	3.0×10^{-2}	
^{240}Pu	1.22×10^{11}	2.25	1.02×10^{3}	
^{242}Pu	7.1×10^{10}	2.18	1.7×10^{3}	
$^{236}PuO_2$				4.3×10^{5}
$^{238}PuO_2$				1.4×10^{4}
$^{239}PuO_2$				45
$^{240}PuO_2$				170
$^{242}PuO_2$				2.7
$^{236}PuF_4$				6.5×10^{7}
$^{238}PuF_4$				2.1×10^{6}
$^{239}PuF_4$				4.3×10^{3}
$^{240}PuF_4$				1.6×10^{4}
$^{242}PuF_4$				1.7×10^{2}
$^{241}AmO_2$				2.6×10^{3}
$^{241}AmF_3$				4.0×10^{5}

[a] Curium isotopes (^{242}Cm, ^{244}Cm) are produced in small quantities in high burnup fuel. However, these isotopes can give rise to significant neutron output arising from their high spontaneous fission and (α, *n*) reaction yields (~10^7 n/s per g).

This equation is valid for chemical separation times ranging from 50 to at least 2100 d and for ^{241}Pu weight fractions ranging from 0.01 to 0.12 at the time of separation.

For MOX, there is a decrease in the dose rate, primarily due to the self-absorption of the low energy photons in the oxide mixture. The MOX surface dose rate equation is:

$$D_{sm}\text{ (mGy/h)} = D_s\{0.13(1 - x)^{0.75} + x^{0.75}\} + 1.0\text{ mGy/h}$$

where

D_{sm} is the surface dose rate of MOX (mGy/h),
D_s is the surface dose rate of PuO_2 (mGy/h),
x is the weight fraction of PuO_2 in the mixture.

In addition, the following restrictions must be applied for the preceding equations to provide valid estimations of the surface dose:

- The sources must be infinitely thick to their own radiations. For plutonium metal this is a thickness of 0.1 cm and for loose oxide powder it is a thickness of 1 cm.
- A shield (rubber glove, plastic container, etc.) of at least 50 mg/cm^2 thickness must cover the source.

The above equations, based on experimental data, are very useful in conjunction with attenuation factors (Table II–1) for estimating potential exposure in problem areas.

II–3. ATTENUATION FACTOR FOR PLUTONIUM GAMMA RADIATION

Table II–1 contains some measured attenuation factors for several shield materials for two isotopic compositions of plutonium

II–4. NEUTRON YIELDS AND DOSE RATE FROM PLUTONIUM

The total neutron yield from plutonium compounds such as PuO_2 includes neutrons from spontaneous fission, (α, n) reactions with oxygen and other light element impurities and induced fissions (neutron multiplication), all of which produce neutrons of different energies. Neutron yields from plutonium isotopes and selected compounds are given in Table II–2 [II–1 to II–3]. A ten energy group neutron source spectrum is given in Table II–3 for mixed oxide fission and the (α, n) source of neutrons [II–4]. Computer codes such as QAD-Pu and ANISN are required for calculating the neutron exposure rates as well as the effectiveness of neutron shields.

For a simplified evaluation of the neutron dose rates, masses of plutonium can generally be treated as a point source of neutrons located at the centre of the mass. The neutron dose rate H (mSv/h) at a distance r (centimetres) from the centre of a large mass of plutonium, plutonium oxide or mixed oxides can be found from the formula

$$H \text{ (mSv/h)} = (9.7 \times 10^{-5})\, S / r^2$$

TABLE II–3. TEN GROUP NEUTRON SOURCE SPECTRA OF Pu AND Pu COMPOUNDS (NORMALIZED TO 1 n/s)

Energy group	Energy upper limit (MeV)	Energy lower limit (MeV)	Flux to dose conversion (mSv/h per n·cm^{-2}·s^{-1})	Pu fission	Mixed oxide fission	Fluoride (α, n)
1	14.91	5.488	1.47×10^{-3}	0.055	0.0288	0
2	5.488	2.466	1.29×10^{-3}	0.2689	0.4654	0.0140
3	2.466	1.653	1.28×10^{-3}	0.1823	0.2269	0.2425
4	1.653	1.108	1.37×10^{-3}	0.1580	0.1022	0.3523
5	1.108	0.608	1.27×10^{-3}	0.1697	0.0906	0.3082
6	0.608	0.111	6.7×10^{-4}	0.1428	0.0742	0.0831
7	0.111	3.355×10^{-3}	2.0×10^{-4}	0.0226	0.0118	0
8	3.355×10^{-3}	2.902×10^{-5}	4.3×10^{-5}	0.0002	0.0001	0
9	2.902×10^{-5}	4.140×10^{-7}	4.3×10^{-5}	0	0	0
10	4.140×10^{-7}	0	3.7×10^{-5}	0	0	0

TABLE II–4. ANNUAL LIMITS ON INTAKE (ALI) FOR INGESTION AND INHALATION BY WORKERS AS RECOMMENDED BY ICRP PUBLICATION 61 [II–5]

Nuclide	Class of compounds[a]	ALI[b] ingestion (Bq)	ALI[b] inhalation (Bq)	DAC[c] (Bq/m^3)
^{236}Pu	W	1×10^5	800	0.35
	Y	8×10^5	700	0.30
^{238}Pu	W	4×10^4	300	0.13
	Y	3×10^5	300	0.13
^{239}Pu	W	4×10^4	300	0.13
	Y	3×10^5	300	0.13
^{240}Pu	W	4×10^4	300	0.13
	Y	3×10^5	300	0.13
^{241}Pu	W	2×10^6	2×10^4	8.6
	Y	2×10^7	2×10^4	8.6
^{242}Pu	W	4×10^4	300	0.13
	Y	3×10^5	300	0.13
^{241}Am	W	3×10^4	300	0.13

[a] Class of compound applies to all isotopes (W: weekly; Y: yearly). Revised classification for W and Y class compounds are M and S, respectively (moderate and slow clearance).

[b] The ALI is based on a committed effective dose of 20 mSv.

[c] DAC: derived air concentration.

TABLE II–5. PROMPT NEUTRON AND GAMMA DOSE AT THE EXTERIOR OF A NORMAL CONCRETE SHIELD FROM A NUCLEAR REACTION OF 10^{18} FISSIONS[a]

Concrete shield thickness (m)	Dose at outer side of shield (Sv)	
	From a nuclear reaction in a metal	From a nuclear reaction in an aqueous solution
0 (0.6 m in air)		980
0.30	880	52.0
0.91	3.17	0.23
1.22	0.17	1.90×10^{-2}
1.53	9.60×10^{-3}	1.40×10^{-3}
1.83	5.90×10^{-4}	1.20×10^{-4}

[a] The dose rate may be calculated for any other number of fissions by a direct proportion.

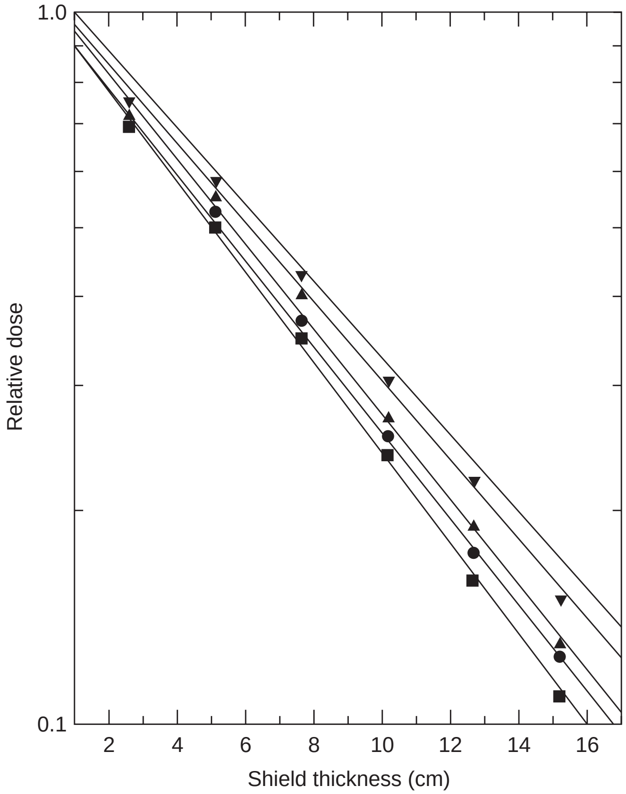

FIG. II–1. Relative reduction in the neutron dose from a PuO_2 *source for different shielding materials (■ : polythene; ● : optigel/water; ▲ : polythene plus B4C; ▼ : WEP-55 acrylic). (The plutonium in the* PuO_2 *source is from high burnup fuel with an average neutron energy of 2.1. MeV. The results were simulated using a* ^{252}Cf *source with a 61* cm^2 *slab shield.)*

where *S*, the total neutron emission rate, is equal to the product of the mass of plutonium (g) times *Y*, the total neutron yield per gram of plutonium (n/s per g) from spontaneous fission, (α, *n*) reactions and fission induced neutrons. Neutron dose reduction as a function of shield materials and thicknesses is shown graphically in Fig. II–1 [II–3].

II–5. INTERNAL EXPOSURE

The radioactive properties and the biological behaviour of plutonium make it a very toxic material when taken into the body, as discussed in Section 5. The major routes of entry for plutonium into the body are:

- By ingestion and subsequent absorption in the GI tract,
- Through a wound,
- By inhalation and subsequent absorption from the lungs.

The annual limits on intake (ALI) for ingestion and inhalation as recommended by the ICRP [II–5] are presented in Table II–4. The derived air concentration (DAC) values for the plutonium compounds are also presented in Table II–4 for application in operational areas.

II–6. RADIATION EXPOSURE FROM CRITICALITY INCIDENTS

Prompt neutron and gamma dose rates at the exterior of an ordinary concrete shield [II–6] from a nuclear excursion of 10^{18} fissions are presented in Table II–5. The

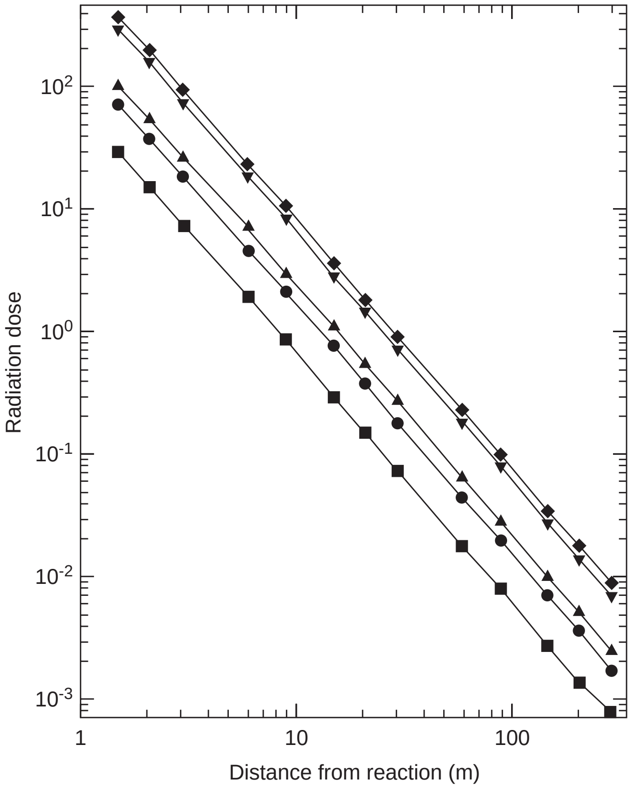

FIG. II–2. Prompt neutron and gamma radiation dose from a burst of 10^{18} fissions as a function of distance from the source (—■— : neutron dose (Gy); —●— : gamma dose (Sv); —▲— : total dose (Gy); —▼— : neutron dose (Sv); —◆— : total dose (Sv)).

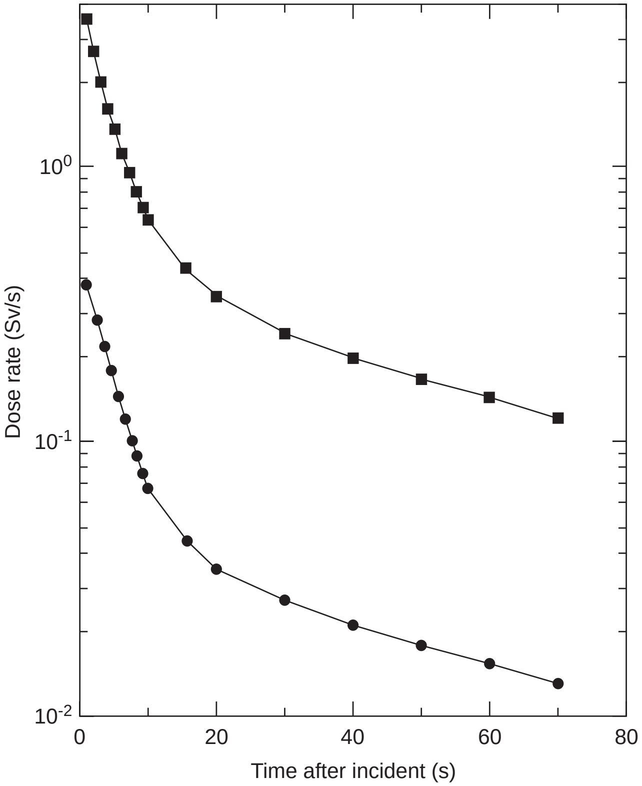

FIG. II–3. Fission product gamma dose rate from 10^{18} fissions (single excursion, unshielded) (the distance from the source is —■— : 3 m; —●— : 9 m).

dose rates in air from the burst in a solution system as a function of distance from the source are shown graphically in Fig. II–2 [II–7].

After the system has become subcritical, the fission products generated in the burst give rise to a gamma dose. The gamma dose rates as functions of decay time at 3 m and 9 m away from the source are shown in Fig. II–3. Time integrated fission product gamma doses are shown in Fig. II–4 [II–8].

The ratio of neutron to gamma radiation dose depends on whether the system is a solution system or a solid one. In a solid system, the ratio can be as high as 10 and for a solution system, it can as low as 0.1.

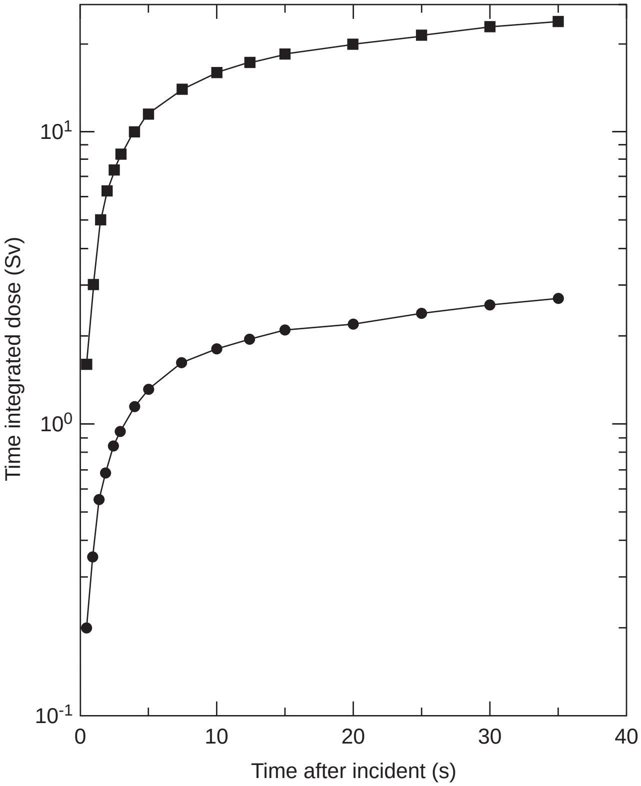

FIG. II–4. Time integrated fission product gamma dose from 10^{18} fissions (single excursion, unshielded) (the distance from the source is —■— : 3 m; —●— : 9 m).

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Annex III

CRITICALITY SAFETY PARAMETERS

III–1. INTRODUCTION

In this annex, criticality safety data are provided for homogeneous aqueous solutions of plutonium, plutonium metal and plutonium compounds, as well as homogeneous mixtures of plutonium and natural uranium [III–1].

III–2. PLUTONIUM SYSTEMS

III–2.1. Aqueous solutions of plutonium

Table III–1 gives the single parameter limiting value (or single parameter subcritical limit) which will ensure that a plutonium nitrate solution system will remain subcritical provided the other conditions are maintained. This subcritical limit allows for uncertainties in the calculations and experimental data used in its derivation, but does not allow for contingencies such as double batching or failure of analytical techniques to yield accurate values. Since the subcritical limits of Table III–1 apply to a uniform aqueous solution of ^{239}Pu nitrate reflected by an unlimited thickness of water without allowances for such contingencies, the process specifications should incorporate a safety margin to protect against uncertainty in the controlled process variable and against the limit being accidentally exceeded.

TABLE III–1. SINGLE PARAMETER LIMITS FOR UNIFORM NITRATE AQUEOUS SOLUTIONS OF ^{239}Pu REFLECTED BY AN EFFECTIVELY INFINITE THICKNESS OF WATER

Parameter	Subcritical limit for ^{239}Pu N: Pu ≥ 4
Mass of ^{239}Pu (kg)	0.51
Solution cylinder diameter (cm)	15.7
Solution slab thickness (cm)	5.8
Solution volume (L)	7.7
Concentration of fissile nuclide (g/L)	7.0
Area density of fissile nuclide (g/cm^2)	0.25
Atomic ratio of hydrogen to plutonium (lower limit)	3630

TABLE III–2. SINGLE PARAMETER LIMITS FOR ^{239}Pu METAL REFLECTED BY AN EFFECTIVELY INFINITE THICKNESS OF WATER

Parameter	Subcritical limit for ^{239}Pu
Mass of ^{239}Pu (kg)	4.9
Cylinder diameter (cm)	4.4
Slab thickness (cm)	0.65
Maximum density for which mass and dimension limits apply (g/cm^3)	19.7

TABLE III–3. SUBCRITICAL MASS LIMITS FOR DRY PLUTONIUM COMPOUNDS

Fissile material form	Density of Pu or compound (g/cm^3)	Subcritical mass limit (kg)
^{239}Pu metal	19.7	4.9
$^{239}PuO_2$	11.4	9.0
$^{239}Pu_2O_3$	11.4	9.0
$^{239}PuF_3$	9.3	10.8
$^{239}PuF_4$	7.0	16.0
$^{239}PuCl_3$	5.7	36.0
$^{239}Pu_2C_3$	12.7	9.96[a]
^{239}PuC	13.6	9.07[a]

[a] Critical mass computed values (see Section III–2.3.)

III–2.2. Plutonium metal units

Table III–2 gives the single parameter limiting value (or single parameter subcritical limit) which will ensure that a plutonium metal system will remain subcritical provided the other conditions are maintained. Water flooding is one assumed contingency. Although flooding is not likely to occur in plutonium handling areas, assuming that flooding will occur (by including the effect of neutron reflection by an infinite thickness of water surrounding the plutonium) introduces a conservative estimate for the unknown water reflecting properties of nearby concrete walls, floors, neighbouring process vessels and transient personnel.

TABLE III–4. MAXIMUM ANNULAR THICKNESS FOR SUBCRITICAL AQUEOUS SOLUTIONS OF ^{239}Pu OF ANY CONCENTRATION

External water reflector thickness (mm)	Annular thickness (mm)
25	63
300	53

III–2.3. Plutonium compounds

Single parameter subcritical mass limits for unmoderated compounds of plutonium (where the hydrogen to plutonium atom ratio H/Pu = 0) at theoretical density under water reflection are listed in Table III–3. The value for plutonium metal from Table III–2 is included for comparison. The limits for reduced density of these compounds can be arrived at by using density scaling or density exponent rules. The values shown in Table III–3 for plutonium carbides are critical masses, computed by the KEN code and are not subcritical mass limits (uncertainties in the calculations are not accounted for).

III–2.4. Annular cylinders

Annular cylindrical tanks formed by two coaxial cylinders with a neutron absorber in its construction may be used for storing plutonium solutions of large volume. The inner cylinder is lined with a cadmium sheet of thickness 0.5 mm and is filled with material with a hydrogen density equivalent to water. Table III–4 gives the annular width acceptable for the storage of aqueous solutions of plutonium. The width of the annulus may be formed by any combination of inner and outer radii. There is no restriction on the concentration of plutonium in the solution or on the solution height.

III–2.5. Soluble and solid neutron absorbers

A significant relaxation of criticality limits can be achieved by the introduction of neutron absorbers in either soluble form or in non-soluble (fixed) form. Soluble poisons are recommended only for secondary protection, as in an auxiliary tank, where fissile material can appear only as a result of incorrect operation. Soluble poisons may be used as a primary means of precluding criticality provided the vessel is behind a massive radiation shield (such as a dissolver). However, it must be based on a fail-safe system of poison addition. Addition of nuclear poison at twice the concentration calculated for $K_\infty = 1$ is generally recommended as a safety criterion.

American National Standard N16.4-1979 [III–2, III–3] provides guidance on the use of borosilicate glass raschig rings as solid neutron absorbers for criticality control in plants processing fissile materials. This standard specifies limiting concentrations of fissile materials in vessels of unlimited size when packed with raschig rings. The recommended limits for aqueous plutonium solutions are presented in Table III–5.

III–3. MIXED (U, Pu) OXIDE SYSTEMS [III–1]

III–3.1. Subcritical limits for ^{239}Pu content

Subcritical limits for the ^{239}Pu content expressed as the weight per cent of $^{239}PuO_2$ in ($^{239}PuO_2 + UO_2$) or ^{239}Pu in (Pu + U) in solutions or aqueous mixtures of

TABLE III–5. MAXIMUM CONCENTRATIONS OF HOMOGENEOUS SOLUTIONS OF PLUTONIUM IN VESSELS OF UNLIMITED SIZE PACKED WITH BOROSILICATE GLASS RASCHIG RINGS[a]

Pu composition	Maximum plutonium concentration (g/L of solution)		
	Minimum glass content in vessel (vol. %)		
	24	28	32
Plutonium containing >5 wt% of ^{240}Pu	140	170	220
Plutonium containing <5 wt% ^{240}Pu	115	140	180

[a] **Specifications:** The density of the glass should not be less than 2.2 g/cm^3 at 25°C and the outside diameter of the rings no greater than 38.1 mm. The boron content of the glass should be between 3.66 and 4.28 wt% boron (11.8–13.8 wt% B_2O_3) and the atomic ratio $^{10}B:^{11}B \geq 0.24$. The density of hydrogen in the solution should be between 75 and 115 g/L, The plutonium should contain at least 50 wt% ^{239}Pu, more ^{240}Pu than ^{241}Pu and no more than 15 wt% ^{241}Pu.

TABLE III–6. SUBCRITICAL CONCENTRATION LIMITS FOR ^{239}PU IN MIXTURES OF PLUTONIUM AND NATURAL URANIUM OF UNLIMITED MASS

Materials	Concentration, Pu/(Pu + U) (wt%)
Dry oxides, H: (Pu + U) = 0	4.4
Damp oxides, H: (Pu + U) ≤ 0.4	1.8
Oxides in water	0.13
Nitrate solutions	0.65

oxides for vessels of unlimited size are presented in Table III–6. The neutron multiplication factor for infinite mass or volume of each of the materials listed in Table III–6 will be less than unity regardless of the density. For example, dry (Pu, U) mixed oxides of infinite mass cannot be made critical if the plutonium concentration does not exceed 4.4 wt% of the total (Pu + U).

III–3.2. Subcritical mass limits

Subcritical mass limits for dry and damp mixed oxides of plutonium and natural uranium for different PuO_2 contents are presented in Table III–7. Data for damp mixed oxide is provided because completely dry conditions may be difficult to maintain. The dampness at an atom ratio H/(Pu + U) = 0.45 corresponds to 1.48 wt% water.

III–3.3. Limiting subcritical concentrations

Subcritical plutonium concentrations for homogeneous aqueous mixtures of the oxides of plutonium and natural uranium are presented in Table III–8. Three different isotopic compositions of plutonium are considered. Plutonium concentration limits are given for 3, 8, 15 and 30 wt% PuO_2 in the total (PuO_2 + UO_2) mixture. When there is less than 3 wt% PuO_2 in the mixed oxide, the subcritical limit of 6.8 g Pu/L in Table III–8 must be reduced to offset the ^{235}U in natural uranium, which becomes relatively more important at lower plutonium content. For example, at 0.13 wt% PuO_2 in mixed oxide, the plutonium concentration limit is 4.9 g/L. Aqueous mixtures having PuO_2 content between 0.13 and 3 wt% PuO_2 must be treated as special cases. If the PuO_2 content is less than 0.13 wt%, criticality is not possible, as noted in Table III–6. The limits of Table III–8 are applicable to aqueous solutions of soluble compounds of (Pu + U), for example nitrates.

TABLE III–7. SUBCRITICAL MASS LIMITS FOR SINGLE UNITS OF MIXED OXIDES OF PLUTONIUM AND NATURAL URANIUM. MASSES GIVEN ARE FOR THE Pu CONTAINED IN THE MIXED OXIDE, AND FOR THE PERMISSIBLE QUANTITY OF PuO_2 + UO_2. THE LIMITS APPLY TO MIXED OXIDES OF ^{239}Pu AND NATURAL URANIUM

PuO_2 in (PuO_2 + UO_2)	wt%			
	3	8	15	30
Dry mixed oxides at theoretical density ≤11.0 g/cm^3				
Mass Pu (kg)	Subcritical in any amount	122	47.0	26.1
Mass of oxide (kg)	Subcritical in any amount	1729	355	98.6
Damp mixed oxides at theoretical density ≤9.4 g/cm^3				
H: (Pu + U) ≤0.45 Mass Pu (kg)	236	49.4	32.9	23.3
Mass of oxide (kg)	8919	700	249	88.1
Damp mixed oxides at one-half density[a] ≤4.7 g/cm^3				
H: (Pu + U) ≤0.45 Mass Pu (kg)	855	161	102	67.9
Mass of oxide (kg)	33 447	2282	771	256.6

[a] Application of these limits requires that the total oxide density does not exceed 4.7 g/cm^3 — i.e. powders.

III–4. SAFETY MARGINS

Safe operating limits are obtained by the application of suitable safety factors to the critical controlling parameters (single parameter subcritical limits). The safety factor should provide allowances for:

- Uncertainty in the value of nuclear constants used and inaccuracies in the method of computation (accounted for in single parameter limits).

TABLE III–8. LIMITING SUBCRITICAL CONCENTRATIONS OF UNLIMITED VOLUMES OF UNIFORM AQUEOUS MIXTURES OF PuO_2 AND NATURAL UO_2

PuO_2 in (PuO_2 + UO_2) (wt%)	3%			8%			15%			30%		
Plutonium isotopic composition[a]	I	II	III	I	II	III	I	II	III	I	II	III
H: Pu atom ratio	3780	3203	2780	3780	3210	2790	3780	3237	2818	3780	3253	2848
Pu concentration (g/L)	6.8	8.06	9.27	6.9	8.19	9.43	7.0	8.16	9.39	7.0	8.12	9.32
(PuO_2 + UO_2) concentration (g/L)	257	305	351	97.8	116	134	52.9	61.7	71.0	26.5	30.7	35.2

[a] Plutonium isotopic composition:
I $^{240}Pu > ^{241}Pu$.
II $^{240}Pu \geq 15$ wt% and $^{241}Pu \leq 6$ wt%
III $^{240}Pu \geq 25$ wt% and $^{241}Pu \leq 15$ wt%

These limits also apply to solutions of plutonium and natural uranium provided all specific conditions are satisfied.

TABLE III–9. RECOMMENDED SAFETY FACTORS [III-4]

Controlling parameter	Mass		Volume	Cylinder diameter	Slab thickness	Concentration
	Batch doubling					
	Possible	Impossible				
Heterogeneity possible	0.43	0.7	0.75	0.85	0.75	0.85
Heterogeneity impossible	0.43	0.85	0.85	0.9	0.85	

- — Inhomogeneities arising owing to varying concentrations, e.g. the presence of a sludge, undissolved particles in solution, non-uniform distribution brought about by a change of state, etc.
- — Difficulties in obtaining a representative and consistent sample, particularly of non-uniform medium.
- — Fluctuations in the accuracy of analyses.
- — Improper operation by plant personnel or of the plant control mechanism, e.g. batch doubling.
- — Other unforeseen circumstances.

Examples of safety factors applied on different controlling parameters are listed in Table III–9 [III–4, III–5]. The single parameter limits of Table III–1 on mass and dimension are valid for any ^{239}Pu concentration in the solution. Safe operating limits obtained from Tables III–1 and III–2, after applying appropriate safety factors for possible contingencies, constitute a set of ‘ever-safe’ parameters for individual systems. Critical dimensions of equipment or critical concentrations can be evaluated for the actual process conditions, including incorrect operations. Safe operating limits are then obtained by applying appropriate safety factors to the controlling parameter such as mass, cylinder diameter, slab thickness or concentration. In such cases, the use of ever-safe parameters would be an unnecessary restriction on the throughputs of a commercial scale plutonium processing plant.

The advantage of using fractional critical masses, volumes or dimensions is the ease of application using readily available criticality data. While these values are satisfactory in most cases, large systems may have a k_{eff} close to unity, the critical condition. On the other hand, the fraction of the critical dimension method may be overly restrictive for smaller systems. For safe operation in plutonium processing plants, this method is accepted in most nuclear establishments (in France, India, Japan, the United Kingdom and the USA).

A safety factor, based on the k_{eff} of a system, may provide a more consistent approach to safety. This method must be used with caution when dealing with small critical systems, where small changes in critical dimensions may result in large changes in the k_{eff}. This approach, based on a safe k_{eff} for the system, is being used for the storage and transport arrays of fissile materials and for the transport of irradiated fuel assemblies. The criticality specialist must consider the limitations on the use of either method in applying safety margins to critical limits.

III–5. STORAGE ARRAYS

An American National Standard Guide [III–6] presents mass limits for spherical units of fissile materials assembled in cubical arrays reflected by ‘thick’ water. The tabulated arrays have a neutron multiplication factor not exceeding 0.95. This standard is directly applicable to the storage of plutonium metal, as well as wet and dry oxides. The water content of the oxides varies between 1.4 and 40 wt% (i.e. an atomic ratio of $0.4 \leq H/Pu \leq 20$). The factors for reducing the mass limits are provided for concrete reflected arrays.

Each unit of the array must remain subcritical if immersed in water. The possibility of double batching of the units in a storage cell should be considered when establishing safety limits and operating procedures. Administrative controls, limited capacity containers and storage cell designs may be useful for the prevention of double batching. Spacing of the units may be maintained by the use of ‘birdcage’ fixtures or other physical barriers.

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GLOSSARY

Definitions of terms that are specific to the handling and storage of plutonium are provided below. No attempt is made to define terms that are routinely used as part of the technical language related to radioactive materials.

absorbed dose. The amount of energy actually deposited in a material or living tissue as a result of exposure to radiation. Units are in rad or gray; 1 gray = 100 rad.

absorption fraction. The fraction of an element which reaches the bloodstream by any mechanism (ICRP Publication 67).

ALARA. As low as reasonably achievable. Personnel radiation exposure shall be maintained as low as reasonably achievable. Radiation exposure of the workforce and the public shall be controlled such that radiation exposures are well below the regulatory limits and there is no radiation exposure without commensurate benefit.

ALI. Annual limit of intake. The amount of material which, if taken internally, will result in limiting the committed dose. ALIs differ depending on the route of intake.

ANISN. A computer code used for neutron transport, criticality and radiation shielding calculations.

barrier. Something that blocks or is intended to block movement. A structure or dynamic effect (e.g. airflow) designed to prevent the transfer of contamination from one area to another.

bird cage fixtures. A framework/open lattice structure designed to maintain separation of fissile material packages for criticality control purposes, while minimizing moderation and reflection.

blister boxes. A temporary extension to a glove box, used to maintain containment during operations (e.g. replacing a glove box window) in which glove box integrity is lost.

burnup. A measure of the consumption of nuclear fuel. Burnup is typically measured in energy extracted per unit mass (gigawatt days per tonne (heavy metal)).

CAM. Continuous air monitor for detecting airborne radioactive contamination.

CANDU. Reactor type designed in Canada using heavy water as a moderator and natural (or low enriched) uranium as fuel.

cells. Space of the facility containing active process equipment and from which personnel are normally excluded.

chelating agents. Chemicals which bind with heavy metals (e.g. plutonium) in blood. These agents are used to enhance the excretion of the heavy metal through urine. Chelating agents are typically administered following an intake of plutonium to minimize the committed dose to the person.

committed dose equivalent (CDE). The dose equivalent calculated to be received by a tissue or organ over a 50 year period after the intake of a radionuclide into the body. It does not include contributions from radiation sources external to the body. The committed dose equivalent is expressed in units of rem (or sieverts).

committed effective dose. The sum of the products of the committed organ or tissue equivalent doses and the appropriate organ or tissue weighting factors (ICRP Publication 60).

committed effective dose equivalent (CEDE). The sum of the committed dose equivalents to various tissues in the body, each multiplied by the appropriate tissue weighting factor. The CEDE is expressed in units of sieverts, as defined on page 26 of ICRP Publication 26.

committed effective dose per unit intake. The committed effective dose expected to be received per unit of activity intake (Sv/Bq). This factor will be different for different routes of intake.

committed equivalent dose. The dose an organ or tissue will receive over the next 50 years. It is a function of the dose of the radiation weighing factor, but does *not* include the tissue weighting factor.

confinement. See ‘containment’.

contain (or confine). To keep within limits; to restrain or control or to hold.

containment. The act or process or condition of containing (or confining).

derived air concentration (DAC). The amount of contamination in air which if breathed for 2000 h/a would result in the annual limit of intake.

derived investigation level (DIL). A value of a bioassay or air monitoring measurement that triggers an investigation.

disposition. Final action (when applied to plutonium, it refers to use or disposal as waste).

disproportionation. A self/auto-oxidation reduction reaction. For disproportionation to occur, an element must have at least three oxidation states and these ions must be able to co-exist in a solution, which in turn is governed by the redox potentials of the couples.

DTPA. Diethylenetriamine penta acetic acid. A chelating agent used to increase the excretion rate of plutonium from the body after an accidental uptake of Pu.

Euratom. European Atomic Energy Community. The agency responsible for nuclear safeguards in the nations of the European Union.

glanded. Sealing provided in a glove box when a cable or pipe passes directly through a wall (or other containment); the gland is compressed plastic material used to provide a seal.

glove box. An enclosure designed to totally confine hazardous/radioactive materials while permitting operations to be undertaken through gloves set into the walls. A glove box is intended to contain plutonium (or other radioactive materials) while it is being processed. The glove box is equipped with gloves, ventilation, filtration, etc.

GM. Geiger–Müller tube. A type of gas filled tube used to detect ionizing radiation.

Halon. The trade name for a series of halogenated (fluro and bromo) carbon compounds used for fighting fires.

high efficiency particulate air (HEPA) filter. A type of filter used in ventilation systems in nuclear facilities (typically 99.97% efficient).

Hypalon. The trade name for a polymeric/plastic coated ambidextrous glove; the coating is chloro-sulphonated polyethylene.

kerma. The quantity K is defined as: $K = dE_{tr}/dm$, where dE_{tr} is the sum of the initial kinetic energies of all charged ionizing particles liberated by uncharged ionizing particles in a material of mass dm. The SI unit for the kerma is the joule per kilogram (J/kg) and is called the gray (Gy).

MAGNOX. Gas cooled reactor. A first generation reactor type in the United Kingdom using a graphite moderator, carbon dioxide coolant and natural uranium metallic fuel.

MDA. Minimum detectable activity.

mean migration coefficient. Measured coefficient quantifying the relationship between the concentration of a species in the soil and its migration rate (cm^2/s).

MERC. Mobile equipment replacement cask.

military plutonium. Plutonium produced for military purposes; may have varying content and be of various grades.

mm WG. Millimetres water gauge. Measures the pressure differential between two areas (10 333 mm WG = 1 atmosphere).

mole (mol). 6.023×10^{23} atoms, molecules or other entities (equal to the number of carbon atoms in 12 g of ^{12}C).

MOX. Mixed oxide (Pu and U oxides) ceramic nuclear fuel.

MSK. A scale by which earthquakes are measured.

non-penetrating radiation. Radiation of such low penetrating power that the absorbed dose from human exposure is in the skin and does not reach deeper organs to any significant, damaging extent. It refers to alpha and beta.

nuclear accident dosimeter (NAD). An instrument for measuring the neutron and gamma radiation of criticality incidents.

penetrating radiation. External radiation of such penetrating power that the absorbed dose from exposure is delivered in significant and damaging quantities to human tissues and other organs. It refers to most gamma radiation, X ray radiation (excluding those with very low energy) and neutron radiation.

pH. Measure of acidity of aqueous solutions; negative logarithm to the base 10 of the H^+ concentration in moles per litre.

posting. A term used in the United Kingdom to describe transferring to and from a glove box. Traditional posting techniques refer to bag-in (or bagging-in) and bag-out (or bagging-out) methods for introducing material into or removing material from a glove box.

PPE. Personal protective equipment.

ppm. Parts per million.

processing. Chemical or physical treatment that alters the properties of a material.

quartz fibre electroscope (QFE). An instrument used for radiation dosimetry.

reprocessing. Separation of Pu, U and fission products from irradiated nuclear fuel.

safe shutdown earthquake (SSE). The most severe earthquake assumed in the design of a nuclear facility.

self-heating. A phenomenon whereby a (radioactive) material heats itself by absorbing the energy that it gives off internally.

STP. Standard temperature and pressure (standard conditions); 0°C and 760 mm of Hg (1 atmosphere).

tri-n-butylphosphate (TBP). Used, usually dissolved in odourless kerosene or dodecane, as a solvent in a chemical reprocessing plant.

tent. A temporary plastic enclosure constructed around a glove box during operations in which the plutonium containment capability of the glove box is lost.

TID. Nuclear safety guide.

TLD. Thermoluminescent dosimeter. An instrument for measuring personnel exposure to radiation.

translocation models. Mathematical models (sometimes referred to as ‘biokinetic models’) which are used to predict the intake, retention and excretion of radioactive material taken into the body.

t/a. Tonnes/annum (1000 kg/a).

weapons grade plutonium. Plutonium containing nominally 93% ^{239}Pu.

zone (or area). Regions or volumes separated by barriers (static or dynamic) which typically contain materials of different risk.

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