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ANALYSIS OF SAFETY TRAINING COURSE

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PLUTONIUM SAFETY TRAINING COURSE

by

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ABSTRACT

This course seeks to achieve two objectives: to provide initial safety training for people just beginning work with plutonium, and to serve as a review and reference source for those already engaged in such work. Numerous references have been included to provide information sources for those wishing to pursue certain topics more fully.

The first part of the course content deals with the general safety approach used in dealing with hazardous materials. Following is a discussion of the four properties of plutonium that lead to potential hazards: radioactivity, toxicity, nuclear properties, and spontaneous ignition. Next, the various hazards arising from these properties are treated. The relative hazards of both internal and external radiation sources are discussed, as well as the specific hazards when plutonium is the source. Similarly, the general hazards involved in a criticality, fire, or explosion are treated. Comments are made concerning the specific hazards when plutonium is involved. A brief summary comparison between the hazards of the transplutonium nuclides relative to ^{239}Pu follows.

The final portion deals with control procedures with respect to contamination, internal and external exposure, nuclear safety, and fire protection. The philosophy and approach to emergency planning are also discussed.

I. INTRODUCTION

Since the discovery of the man-made element plutonium in 1941, the importance and usefulness of this element has steadily increased.¹⁻³ In addition to its application in nuclear weapons, plutonium is an important source of nuclear fuel for the production of electric power in reactors. Lightweight plutonium-fueled power units produce electricity to run devices such as pacemakers and space experiment packages. Plutonium is used with certain other materials to produce a source of neutrons. This element is also useful in the detection of neutrons. The element is used as a source material for the production of other substances called transplutonium elements. A number of these elements have already been produced.

The first plutonium was produced from uranium in a cyclotron. Since then, much greater amounts have been produced from uranium used in reactors. Reactors using uranium of low enrichment as fuel produce large amounts of plutonium. However, much of this plutonium is also used up as the reactor fuel before it can be recovered. The net content of plutonium in discharged fuel runs about 0.5-1.0%. This can then be recovered for reuse when the spent fuel is reprocessed.⁴ Since existing power reactors generate so much plutonium, the recycling of this plutonium for use as the fuel of the power reactor is an attractive concept.

The special features of plutonium that increase its utility require that this substance be handled most carefully. This need for safety arises from four main features of the element. First, plutonium is an unstable substance, whose decay results in the release of radiation. This radiation may present a hazard, even if one is distant from the source. Second, the element is toxic. Intake into the body may result in long-term deposition in bone with severe local damage at these sites. Third, the substance is fissile. When enough of this material is present in the proper setting, a fission chain reaction can occur. Such an event would release large amounts of energy and radiation. Fourth, some forms of plutonium are pyrophoric; that is, they ignite spontaneously. Once started, a fire could spread rapidly and result in great damage. To reduce the hazard potential and to achieve safety in handling plutonium, control of these features is required.

A. General Safety/-design Approach

Through the years, the safety-design philosophy for plutonium has evolved into the central theme "confinement and containment." Confine the process to ensure a barrier exists between the worker and the toxic substance. The barrier then protects the worker from the hazards of the material. A source enclosed in a capsule would be a simple means of confining or enclosing the material. Sometimes, both the substance and the process need to be confined, and enclosures such as hoods, gloveboxes, hot cells, and other structures are used.⁵⁻¹³

Containment refers to preventing the release of a confined substance. A release should not result in the discharge of a hazardous amount of the confined substance from the facility.¹⁴ This requires the presence of other systems to contain any release of confined material when an accident occurs. Proper design of airflow patterns and tandem exhaust filtering provide a multiple barrier to releases. Airflow patterns are directed from clean areas toward those in which hazards may develop, as shown in Fig. 1. Air in these areas flows from offices through corridors into the laboratory to pass through the enclosed system. The enclosure (hood, glovebox, etc.) atmosphere is discharged through the filter system. The flow pattern is always into the enclosure, which reduces the chance of material escaping. The room, the airflow, and the filter system provide the containment. Other more detailed ventilation systems may also be devised.¹⁵⁻¹⁶

Fig. 1. Confinement and Containment for a Plutonium Laboratory

B. Safety-design Philosophy

The purpose of the safety design in all cases should be to achieve these goals:

1. Ensure that personnel radiation exposure is kept as low as reasonably achievable under normal operations.
2. Control effluent release to ensure no increase in present environmental background levels occurs. Ensure that any radiation exposure to nearby population groups is kept as low as reasonably achievable. Design systems to prevent the release of hazardous levels of material in the event of accidents.
3. Include features that will prevent or reduce the severity of incidents. The impact of the event in regard to personnel injury and threat to life should be the prime concern. Damage to property and program delays should also receive some thought.¹⁴

Because of the special hazards that are present in work with plutonium, many design features must be treated. The type of functions to be carried out will affect the degree of safety needed. Basic studies of the site, plant layout, ventilation patterns, waste-handling needs, fire-control approach, radiation-safety problems, and nuclear-safety aspects will dictate many design goals.^{7,17-20} Structure design, process flow, confinement, and containment through gloveboxes or other devices will also call for a certain level of engineered safety.^{11,12,19,20} The form, amount, and composition of the plutonium handled are also factors. These aspects modify the extent of hazards expected from the radiative, biological, nuclear, and ignition properties of plutonium.

C. Operational Safety Measures

The design of safety features into the plant or process is but part of the effort needed to attain safety. Since workers must still perform certain required tasks, human error can be a factor that cancels any gain made by built-in safety. For this reason, a proper administrative approach to safety is crucial. Policy must require and promote safety as an integral part of proper work methods. Such an approach demands that instructions, procedures, or manuals stating the correct sequence and safe methods of performing the work be prepared in detail.^{5,7,12,21} Responsibility for safety, as well as work responsibilities, should be pointed out at each level of job function. Included in the writeup should be safety procedures in regard to waste disposal, fire control, accident prevention, initial decontamination, radiation safety, and emergencies.^{5,7} The material should be reviewed and updated on a frequent basis.

Employees should be instructed in the proper procedures and made aware of their safety responsibilities. The worker has primary responsibility for his own safety, since he performs the work. The worker should be well-trained not only in regard to proper work methods, but also in regard to safety measures. Frequent review and training in regard to safety aspects should be provided. Drills and reviews of the proper action to be taken during emergencies should also be conducted.

The employee should be made aware of whatever hazards are involved in the process. He should be trained in measures to avoid fires, accidents, explosions, and injuries. Workers should know what emergency plans exist and the functions of the emergency groups. They should be instructed in how to obtain help quickly from the proper support group. Training should cover the alarm systems, what they signify, and the proper response to the alarms.

D. Health-physics Measures

Even though design seeks to prevent accidents, and work is done in a safe manner, one cannot overlook the potential for accidents to occur. These events may result in the release of radioactive matter. Thus safety measures should be backed up by an active health-physics program.

Enclosing the process excludes the toxic substance from the worker's environment. This measure protects him from the toxic effects that result from intake into the body. By proper shield design, external radiation exposure to the worker can be controlled. However, when a breach in the system occurs, this control is lost and serious hazards may result.

The term "contamination" may be used to denote the presence of unwanted radioactive matter. The control of the spread of contamination is one of the functions of the intact enclosure. Alarm devices that respond to a preset

radiation level or to the presence of radioactivity in air denote on a gross scale when contamination control is lost. A health-physics program provides personnel and area-monitoring services that help control the spread of contamination. Frequent surveys of an area or personnel with instruments allows early detection of any contamination. Air sampling allows small amounts of radioactivity to be found. These measures point out small defects in the control system before major hazards occur. In this way, the spread of contamination is limited and measures to locate the defect can be undertaken.

Other aspects of a health-physics program will be discussed later. These include such things as personnel monitoring, bioassay programs, instruments, surveys, remote monitors, and emergency planning.

E. Safety Achievement

A certain degree of safety can be designed into a plant or process. Over and above this, safety practices can be written up, presented, and promoted. Workers can be instructed, trained, and retrained in safe practice and in recognizing hazards. A health and safety program can augment these features by maintaining the safety control achieved by these measures. In the final analysis, the achievement of safety requires a balanced effort which involves the planning and cooperation of many groups. When the concept that safety is a shared responsibility is accepted, then the goal of a safe program can be realized.

II. PROPERTIES OF PLUTONIUM

Although more research has been devoted to methods of producing and using plutonium, much knowledge of the properties of this substance has also been collected.^{2,7,22,23} Plutonium, an unstable element of atomic number 94, is a heavy metal. This substance (given the symbol Pu) is artificially produced during fuel burnup in reactors. The metal has many unique properties. Some of these affect the degree of hazard involved in plutonium work. Some make this substance very hard to work with under certain conditions. Others make this metal very useful. No other metal has such diverse properties. In this section we discuss some of these features to provide a background for later treatment of plutonium hazards.

A. Chemical and Physical Properties

Plutonium metal, which has a bright, silverlike appearance, quickly tarnishes in air.² It corrodes faster in moist air than in dry air. Plutonium is highly reactive. It combines with oxygen to form PuO_2 , PuO , and Pu_2O_3 , reacts with some acids, and forms stable compounds with most nonmetallic elements. The metal exists in six distinct crystal forms (phases) within certain temperature ranges. At room temperature, the metal is normally in the alpha phase. Upon being heated, the metal will expand in some phases and contract in others. When unalloyed, it is hard and brittle; as an alloy, it may become soft and ductile. Although a metal, it is not a good conductor of heat or electricity as most metals are. Because of its radioactive decay, it will self-heat, and this may damage the metal or compound. As decay occurs, a change in the chemical makeup takes place as the plutonium changes to other substances.² Table I, adapted from Ref. 7, contains a brief summary of some of the features of this metal.

TABLE I. Physical and Chemical Properties of Plutonium^{2,7,22}

Density	15.9-19.9 g/cm ³ , depends on metal phase.
Melting point	
Pure metal	640°C.
Alloys	Up to 2000°C, varies with alloy.
Boiling point (pure metal)	3327°C.
Oxidation rate	Slow, in dry air. Rapid under moist conditions or when heated. May result in a low spontaneous ignition temperature.
Action of acids and bases	Dissolves readily in concentrated hydrochloric, hydroiodic, and perchloric acids. Attacked by most dilute acids; not readily attacked by concentrated sulfuric and nitric acids or sodium hydroxide solutions.

B. Radiologic Properties

1. Introduction

The basic structure of all matter consists of the union of one or more of the atoms of certain elements. Air, water, and salt are matter composed of more than one element. Lead, silver, and gold are matter composed of only one element. Each element has a symbol by which a compound may be expressed. Plutonium dioxide may be written PuO_2 ; that is, one atom of plutonium (Pu) and two atoms of oxygen (O).

Of the more than 100 known elements, 90 have been found in nature and the rest have been artificially produced. Some natural elements and all man-made ones are unstable. This means that these substances undergo changes to become other elements until a stable form is reached. This change may be quite rapid or may require millions of years to achieve the final stable state.

2. Atomic Structure

The changes in unstable elements reflect adjustments in the structure of their atoms. An atom is thought to consist of a nucleus surrounded by enough electrons to produce no net charge. The nucleus is the massive, central, positively charged portion of the atom, composed of neutrons and protons (see Fig. 2). The neutron and the proton are particles of about equal mass. The neutron is electrically neutral; the proton has a unit positive charge, and the electron carries a unit negative charge. The structure of an atom of an element may be denoted by ${}^A_Z\text{X}$, where X stands for the element symbol. The mass number, A, gives the total number of neutrons and protons in the nucleus. Z is called the atomic number and gives both the number of protons (or positive charges) in the nucleus and the number of electrons (or negative charges) outside of the nucleus.

The neutron number is obtained by subtracting Z from A. On the basis of atomic weights, both the neutron and the proton have about unit mass, so the mass number is the closest whole number to the atomic weight. The small electron mass adds little to the atomic weight.

The chemical nature of any element is a function of its atomic number. If the Z number changes, the substance becomes some other element denoted by the new Z number. However, the same element may have atoms that differ in mass number. The term "isotope" is used to denote atoms with the same atomic number but with different atomic mass. Since Z is the same, isotopes have equal numbers of protons, but the number of neutrons varies.

The stability of an atom is related to the mass of that atom. The lighter elements, in which the neutrons and protons are about equal, are usually stable. For those of larger mass, an excess of neutrons is required for an atom to be stable. For very large atoms ($Z > 84$), the mass seems to become too large to maintain a stable state. When this occurs, all the isotopes of a given element will be unstable and will adjust their structure until a stable state is attained.

3. Radioactive Decay

To alter the structure of an unstable atom requires a change in the composition of the nucleus. This can be done if the atom undergoes a change in the number of neutrons or protons. One way in which the atom brings about this change is called radioactive decay. In this decay or disintegration process, the nucleus of an unstable atom spontaneously emits certain radiations, which alter the nuclear structure. For a given substance, the rate of decay of its atoms is characteristic of that isotope of the element. The half-life, which expresses the time for one-half of the atoms in a given sample to decay, is related to the rate of decay.

4. Types of Radiation

The term "natural radioactivity" is applied to unstable or radioactive substances that are found in man's natural environment. These emit one or more of three types of radiations: alpha, beta, and gamma rays. Radioactive substances can also be produced by artificial means, such as reactor irradiation, accelerator bombardment, and other ways. These processes also produce other radiations: X rays, positrons, and neutrons.

Radiations are emitted with various amounts of energy, which can be related to the specific source. As they move through matter, this energy will be lost. The rate of loss depends on the type of emission, the nature of the interaction, and the kind of substance involved. The transfer of the energy to matter results in the release of electrons from the atoms of the substance. The process whereby electrons are removed from a neutral atom is called ionization. For this reason, the radiations mentioned above are called ionizing radiations. Sometimes, no electrons are removed during energy transfer to the atom and the process is called excitation.

a. Alpha Radiation. Alpha particles (α) are helium nuclei (${}^4\text{He}$) emitted with a discrete energy and a characteristic half-life of emission. Most naturally occurring alpha sources (atomic weight > 200) emit particles in the energy range 4-9 MeV. (The MeV is a useful energy unit used in nuclear physics and stands for million electron volts.) Alpha particles lose energy mainly by direct ionization and excitation. An alpha particle carries a double positive charge and has a relatively large mass so that it forms a large number of ions along its track and hence quickly loses energy. An alpha particle

is easily absorbed by a few sheets of paper or the outer (dead) skin layer of the body. For alpha particles from natural sources, the range in air is only a few inches.

b. Beta and Positron Radiation. Beta particles (sometimes called negatrons) and positrons are similar and possess many of the same features:

(1) Beta particles (β^- , e^-) are electrons emitted at high speed from the nucleus of the radioactive atom. The electron is a small particle (mass about 1/7300 of that of an alpha particle) with a single negative charge. Beta particles are emitted with a continuous energy spectrum up to some maximum energy, which is characteristic of that source. Natural beta sources emit particles with maximum energy up to about 4 MeV. Low-energy beta particles lose energy mainly through direct ionization and excitation. Some of the energy loss may be through a conversion process called bremsstrahlung (braking radiation), which gives rise to X rays. This conversion process becomes increasingly important as the energy of the beta particle increases and may be the major process at high energies.

Because of the single charge and small mass, beta particles lose energy less rapidly than alpha particles. Ionization by beta particles is roughly 100 times less than that for alpha particles. Beta particles have ranges up to many feet in air and will penetrate a few millimeters of body tissue. X rays are produced in stopping beta particles, and these may also lose energy in the medium.

(2) Positrons (β^+ , e^+) are positive electrons having the same rest mass as an electron but with a positive charge. They are emitted from certain artificially produced radioactive substances with a continuous energy spectrum up to some maximum energy characteristic of the source. Positrons and electrons may also result when high-energy gamma rays interact with matter. The positron loses its energy by the same processes that electrons do. However, a positron that has lost its energy of motion exists for only about 10^{-9} sec. It then interacts in a process called annihilation, which releases a pair of gamma rays. The gamma rays may then also lose energy in the medium.

c. X and Gamma Rays. X and gamma rays are electromagnetic radiation similar to light, but of shorter wavelength. They are called photons and possess similar properties. A photon has an effective momentum, but no mass or charge. Gamma ray is a term usually applied to photons emitted from the nucleus, X rays to those produced outside the nucleus of the atom.

The gamma rays that often accompany alpha- and beta-particle emission are emitted with discrete energies. This energy indicates a transition of an excited nucleus from one level to a more stable one.

X rays (bremsstrahlung) produced when high-energy charged particles are stopped in matter have a spectrum of energies, the spectrum varying from zero up to the particle energy. Characteristic X rays (discrete energy) are emitted when an excited atom returns to a normal energy state.

Photons lose energy to matter by three main processes: photoelectric effect, Compton effect, and pair production.²⁴ Other processes such as photofission and nuclear reactions occur only for very-high-energy photons. In the three main processes, the photon is either absorbed or scattered and energetic electrons are released. These secondaries (e^- and e^+) produce most of the ionization that results in the substance. Thus, X and gamma rays ionize in an indirect manner.

X and gamma rays can travel great distances in a medium before losing any energy. As the photon proceeds onward, it loses no energy until an interaction takes place. The chance of a photon interaction along its path remains constant until one occurs, but only a fraction of all available photons interact at any given point. The rest of the photons move on without any energy loss. The discontinuous feature of photon-energy transfer gives X and gamma rays great penetrating power. Even in dense media, such as lead, the penetration is significant. Photons can enter deeply into the body or even pass through it without interacting.

d. Neutrons. Neutrons (1_0n) are uncharged particles of atomic weight slightly greater than a proton or hydrogen nucleus (1_1H). They are found in the nucleus of every element heavier than hydrogen and in all but one hydrogen isotope. Outside of the nucleus, neutrons are unstable and are transformed to protons after beta decay.

Neutrons are not emitted from natural sources. They result from fission in reactors and are emitted when certain heavy elements undergo spontaneous fission (see Sec. 7 below). Accelerators produce neutrons when high-energy particles strike suitable targets. Radioactive sources, combined with the proper target substance, yield neutrons through reactions of the target nucleus and the emitted radiation. These are referred to as (α, n) or (γ, n) reactions. The fission process and (α, n) reactions produce neutrons with a spectrum of energies. Accelerator and (γ, n) processes yield nearly monoenergetic neutrons.

As they traverse matter, neutrons lose energy through nuclear interactions with the member elements of the media. A number of processes may take place: elastic scattering, inelastic scattering, and absorption leading to radiative capture, fission, charged-particle reactions, or $(n, 2n)$ reactions.^{24,25} In those processes, neutrons, charged particles (recoil nuclei or emitted particles), and gamma rays are released. The ionization that takes place results from the charged particles; thus, neutrons also ionize indirectly, as did X and gamma rays.

As with X and gamma rays, neutrons can penetrate to great depths in a substance. Neutrons lose no energy until one of the competing processes occurs at some point. This means that neutron energy transfer is a discontinuous process as was the case for photons. Since energy is not lost continuously along the path, neutrons can enter deeply into the body or may even pass through without being absorbed.

It is customary to categorize neutrons in terms of the energy ranges in which certain effects predominate. The NCRP²⁶ uses the following scheme: thermal (0.025 eV), intermediate or resonance (1.0 eV to 10 keV), and fast (>10 keV).

5. Plutonium Formation

In Sec. 3 above, it was mentioned that the structure of an unstable atom can be altered by changing the number of neutrons or protons. In radioactive decay, the atom ejects matter, so that the trend is to reduce the size of the nucleus. On the other hand, changes in the nuclear structure may also be caused by adding neutrons or protons.²⁷ Such was the manner in which plutonium first came into being.^{2,27} The unstable element uranium was bombarded by deuterons (${}^2_1\text{H}$) to form a new substance:

In this case, the deuteron enters the uranium nucleus, giving an already unstable atom much more than it can handle. The new excited nucleus "spits" out two neutrons, leaving what remains a new element, ${}^{238}_{93}\text{Np}$. However, this new element is also unstable and decays by beta emission:

Shortly after this first isotope of plutonium was found, the isotope ${}^{239}\text{Pu}$, which is valuable in the nuclear-weapons program, was produced.^{1,2,27} Although the cyclotron was used to produce the first plutonium, this method could not be used to make large amounts of the metal. A method that would supply useful amounts of the substance was developed when the first self-sustained chain reaction was achieved.

In the early reactors, natural uranium, composed mostly of ${}^{238}\text{U}$ with about 0.7% ${}^{235}\text{U}$ and 0.005% ${}^{234}\text{U}$, was used as the fuel. The ${}^{235}\text{U}$ isotope was the actual fuel that kept the chain reaction going. However, neutrons released in the fission or splitting of ${}^{235}\text{U}$ could be captured by the ${}^{238}\text{U}$. This capture process changes ${}^{238}\text{U}$, which decays very slowly, to ${}^{239}\text{U}$, which decays quickly. The product of ${}^{239}\text{U}$ beta decay is ${}^{239}\text{Np}$, which also beta decays fairly quickly to form ${}^{239}\text{Pu}$. In this manner, the supply of ${}^{239}\text{Pu}$ could be built up, since the half-life of ${}^{239}\text{Pu}$ is about 24,400 years.

Not only is the 239 -isotope formed in the reactor, but other plutonium isotopes are also produced (see Fig. 3). Further capture of neutrons converts some of the ^{239}Pu to ^{240}Pu , ^{241}Pu , or even higher isotopes. The relative amounts of the isotopes formed depend upon the type of reactor, the length of irradiation time, and the composition of recycled fuel.⁷ With the advent of power reactors fueled by plutonium, the radiologic properties of the spent fuel are subject to the influence of more isotopes than ^{239}Pu .²⁸⁻³⁰ Figure 4 shows an estimate of how the plutonium isotopic composition varies with burnup in uranium-fueled boiling water and pressurized water reactors. Reference 28 contains estimates of plutonium isotopic composition from plutonium-fueled reactors and a liquid-metal fast breeder reactor.

Fig. 3. Formation of Plutonium Isotopes and Daughters (Adapted from Refs. 28 and 29)

Fig. 4
Plutonium Isotopic Composition as
a Function of Uranium Burnup⁴

The largest amount of plutonium has been produced in reactors, both research and power types. Some plutonium, as well as transplutonium substances, has been produced in accelerators and also has resulted from thermonuclear-weapons tests.

6. Radioactive Decay of Plutonium

The unstable nature of some elements reflects the failure of the forces that bind neutrons and protons in the nucleus. Since like charges repel, protons repulse each other. To balance this, it is assumed that neutrons and protons are attracted by short-range forces. As the mass of the atom increases, though, more neutrons are needed to overcome the repulsive force. If the ratio of neutrons to protons becomes too great, the atom will also be unstable and will favor beta decay. If the ratio is too low, positron decay would be expected.

In the heavy elements with the large content of protons, the degree of instability is such that alpha decay begins to occur. That is, the nucleus begins to eject larger particles in an attempt to reach a more stable structure. As the mass numbers and proton content go even higher, the imbalance in the forces increases.

All 15 known isotopes of plutonium (mass numbers 232-246) are radioactive. Most of these decay by alpha emission. However, ^{241}Pu is a notable beta emitter. For the most part, only the isotopes of mass numbers 238-242 and some of their decay or daughter products need be discussed here. In this respect, the products ^{237}U and ^{241}Am require attention, since they increase the amount of gamma radiation as the plutonium sample ages. These products are the only ones from the decay of the 238-242 group with half-lives short enough to affect the sample activity.

A number of compilations of the decay features of plutonium isotopes and their daughter products have appeared.^{7,22,29-35} Table II, adapted primarily from Ref. 30 and updated, summarizes some of these features.

7. Spontaneous-fission Neutron Radiation

For the heaviest elements, the atoms became so unstable that still additional processes act to change the structure to a more stable one. The unstable atom may split into two large fragments in a process called fission. Since the atom breaks apart in a random process much like radioactive decay, the process is termed "spontaneous" fission. As with radioactive decay, one can assign a half-life for spontaneous fission. The chance of spontaneous fission occurring increases as the Z number increases, and those atoms with even Z and even A seem to have the shortest spontaneous fission half-lives.³⁹

TABLE II. Decay Features of Selected Isotopes^{10,15-17}

Isotope	Half-life ^a	Specific Activity, dis/min-g	Emissions ^b			
			$E_{\alpha,\beta}$, MeV	Type %	$E_{X,\gamma}$, MeV	%
²³⁷ U	6.75 d	1.81×10^{17}	0.245	β^- 80	0.026	2
			0.09	β^- 12	0.060	36
					0.165	2
					0.208	23
					0.267	0.76
					0.332	1.4
					0.37	0.17
²³⁸ Pu	87.8 yr	3.80×10^{13}	5.50	α 72	0.017	13
					0.043	3.8×10^{-2}
			5.46	α 28	0.099	8×10^{-3}
					0.150	1×10^{-3}
					0.77	5×10^{-5}
²³⁹ Pu	2.439×10^4 yr	1.36×10^{11}	5.16	α 88	0.017	3.9
			5.11	α 11	0.039	2×10^{-3}
					0.052	7×10^{-3}
					0.124	2.5×10^{-3}
					0.384	1.5×10^{-3}
					0.414	1.2×10^{-3}
					0.65	8×10^{-5}
					0.77	2×10^{-5}
²⁴⁰ Pu	6.54×10^3 yr	5.06×10^{11}	5.17	α 76	0.017	10
					0.044	1×10^{-2}
			5.12	α 24	0.104	
					0.160	
²⁴¹ Pu	15 yr	2.20×10^{14}	0.021	β^- 99+		
			4.90	α 1.9×10^{-3}	0.145	1.6×10^{-4}
			4.85	α 3×10^{-4}		
²⁴² Pu	3.87×10^5 yr	8.48×10^9	4.90	α 76	0.017	10
			4.86	α 24	0.044	1×10^{-2}
²⁴¹ Am	433 yr	7.61×10^{12}	5.48	α 85	0.017	37
					0.026	2.5
			5.44	α 12.6	0.043	6×10^{-2}
			5.38	α 1.7	0.060	36
					0.101	4×10^{-2}
					0.208	6×10^{-3}
					0.335	8×10^{-4}
					0.370	4×10^{-4}
					0.663	5×10^{-4}
					0.772	3×10^{-4}

^aHalf-life values were obtained from The Chart of the Nuclides, April 1972 edition.^bFigures for the X-ray abundance (0.017 MeV) listed for ²³⁹Pu and ²⁴¹Am were obtained from Ref. 39. For a more detailed listing of X and gamma rays, see Ref. 38.

In the fission process, both fast neutrons and gamma radiation are emitted. The number of neutrons per fission is a function of both the atomic weight and the atomic number of the isotope. The neutrons which are emitted in this process will add to the total radiation which must be dealt with. The rate at which these neutrons are emitted will be a function of the isotopic composition of the plutonium. Table III gives information in regard to the spontaneous fission aspects of some isotopes.

TABLE III. Spontaneous-fission Characteristics

Isotope	Spontaneous Fission ³⁶ Half-life, years	Neutron Emission, ⁴⁰ avg n/fis	Calculated Emission Rate, η/ρ , n/sec-g
²³⁸ Pu	4.9×10^{10}	2.04	2.32×10^3
²³⁹ Pu	5.5×10^{15}	2.15 ^a	0.02
²⁴⁰ Pu	1.34×10^{11}	2.23	9.22×10^2
²⁴² Pu	7.1×10^{10}	2.32	1.79×10^3
²⁴¹ Am	2.0×10^{14}	2.3 ^a	0.62

^aNot from Ref. 40.

8. Neutron Emission from Compounds

Neutrons produced from plutonium by spontaneous fission may not be the only source of neutrons to contend with. The alphas emitted from plutonium are all emitted with rather high energy (see Table II). These alpha rays are able to enter the nuclei of light elements such as aluminum, beryllium, lithium, oxygen, fluorine, and sodium. Such a process results in a compound product which is in an excited or unstable state. This compound then emits a neutron in an effort to return to the ground or stable state for the new element. These (α , n) reactions result in fast neutrons. The neutron yield and the maximum neutron energy for (α , n) processes are a function of the alpha energy and the nature of the target material. There is no simple method by which one can arrive at either the neutron energy or yield.

Although exact neutron-emission rates from plutonium compounds are not well known, some useful estimates have been made.^{30,41} Neutron emission from certain compounds should not be neglected, since they can greatly increase the neutron problem.²⁹ Table IV lists the rates calculated by Steindler³⁰ for plutonium tetrafluoride and dioxide.

TABLE IV. Fast-neutron Emission from Selected Compounds

Compound	(α , n) Emission Rate, n/sec-g Pu
$^{238}\text{PuF}_4$	2.1×10^6
$^{239}\text{PuF}_4$	4.3×10^3
$^{240}\text{PuF}_4$	1.6×10^4
$^{242}\text{PuF}_4$	1.7×10^2
$^{238}\text{PuO}_2$	1.4×10^4
$^{239}\text{PuO}_2$	45
$^{240}\text{PuO}_2$	1.7×10^2
$^{242}\text{PuO}_2$	2.7

C. Nuclear Properties

In the section above, our attention was directed to the unstable nature of plutonium isotopes. For spontaneous fission and decay, the events are random, not subject to outside influences. The (α , n) processes, however, show that the radiations from plutonium can affect the nuclei of other atoms. In this section, we will discuss some of the processes that bring about certain effects in plutonium atoms. For the purposes of this discussion, only the actions of neutrons on the nuclei need be treated.

1. Neutron Reactions

A number of neutron interactions with matter were mentioned in Sec. II.B.4.d. These types of interactions also occur in plutonium. Some of these processes are more likely to occur with slow neutrons; others are more likely to occur with fast neutrons. Some may hardly occur at all in certain energy ranges, and others may occur to some degree regardless of energy range. For slow neutrons, the three reactions--elastic scatter, radiative capture, and fission--are those most likely to occur. In the fast-neutron region, the processes of inelastic scattering and (n, 2n) reactions become possible. Of all these processes, those of capture and fission are of main concern. Radiative capture, i.e., the (n, γ) reaction, results in the buildup of the higher isotopes of plutonium in a reactor (see Fig. 3). Because the fission process takes place in plutonium, this substance can be used as the fuel in power reactors.

2. Fission

The fission process was first detected in the isotope ^{235}U and resulted from attempts to produce a transuranic element.^{24,25} In studies using

neutrons to bombard uranium, slow-neutron capture led to beta emitters that were difficult to identify. In the process, a thermal neutron, n_{th} , is captured by the ^{235}U nucleus (see Fig. 5). This produces a compound nucleus which is

Fig. 5. Fission Process in ^{235}U due to a Thermal Neutron

in an excited state. The result is that the compound nucleus fissions or breaks apart and the two fission fragments carry off most of the energy given up in the process. At the time of the fission, gamma rays, beta particles, and neutrinos (ν), are emitted, as well as neutrons. The beta particles and gamma rays are referred to as prompt. The neutrino is a particle of zero mass which carries off some of the released energy. About 9 MeV/fission of prompt beta particles and about 7 MeV/fission of

prompt gamma rays are emitted in ^{235}U fission.⁴⁰ The fission fragments come off with velocities of about 10^9 cm/sec and may have ranges up to 1 in. in air. Since these fragments have too many neutrons to be stable, they eject one or more neutrons. Most of the neutrons (>99%) are emitted at fission or slightly after the fission occurs; these are known as prompt neutrons. These neutrons are emitted almost isotropically with a spectrum of energies from 0 to 17 MeV. The average energy is about 2 MeV. Some neutrons (called delayed neutrons) are released over a period of minutes. The delayed neutrons are emitted at times $>10^{-3}$ sec after the atom splits. For ^{235}U fission, about 2.5 neutrons are emitted per fission, on the average.

During the decay phase of the fission fragments and resulting fission products, more beta particles and gamma rays are also given off until stable nuclei are formed. The entire fission process results in a release of about 200 MeV/atom fissioned. Of this, fission fragments carry off about 165 MeV. Note that this is the energy released in only one fission. This implies that we can obtain a tremendous amount of energy from just a small amount of uranium because it would contain so many atoms. The energy released in the fission of 1 g of ^{235}U is equivalent to about 21 tons of TNT.^{24,25} The large amount of energy released per fission makes this process very useful as an energy source.

We can distinguish between those substances that can be fissioned and those in which a chain reaction can be sustained. Under the proper conditions, fission has been produced in many substances with $Z \geq 73$. The release of neutrons in the fission process is a vital feature necessary to allow a chain reaction to proceed. The released neutrons may in turn strike other nuclei and lead to more fissions, etc. However, before the chain reaction can maintain itself, a certain amount of the fissile substance must be present. Other processes compete with fission events for neutrons; therefore the supply of neutrons must be large enough to keep the chain going. Part of the reason is that many neutrons are lost while being slowed to thermal energy.

This effect depends upon other substances in the system. Those that slow down neutrons without absorbing them are called "moderators." Those that tend to absorb neutrons are called "poisons." The amount of fissile material needed to sustain fission is called the critical size or mass. For any system the process will be self-sustaining when the number of neutrons lost is balanced by the number produced. In many substances, though, the fission process cannot be made self-sustaining. For this reason, only ^{233}U , ^{235}U , ^{239}Pu , and ^{241}Pu , in which the chain reaction can be maintained, are of value for present thermal-reactor use.

The fission process is not restricted solely to thermal-neutron captures in fissile substances. Some of these materials may be fissioned by fast neutrons or other agents as well; e.g., ^{238}U is fissioned by fast neutrons, but not by thermal neutrons. In Sec. II.B.7 we saw that some substances even undergo spontaneous fission.

3. Fission in Plutonium

All the odd-mass isotopes of plutonium can be fissioned by thermal neutrons, but the short half-life of some of these substances limits their practical value. In essence, ^{240}Pu and ^{242}Pu are nonfissile at low neutron energy. For even-mass products of $A < 240$, thermal fission becomes more probable. The probability of fission at any neutron energy may be expressed by the cross section, σ_{fission} , stated in units called barns. The higher the probability for fission to occur, the higher will be the value of the cross section in barns. The cross-section concept also applies to other processes which compete with fission. Table V gives values of the cross sections for thermal neutrons (0.025 eV) for those processes likely to occur in plutonium isotopes.

TABLE V. Thermal Cross Sections of Plutonium Isotopes⁴²

Isotope	$\sigma_{\text{absorption}}$, barns	σ_{fission} , barns	σ_{capture} , barns	Neutrons/fission
^{239}Pu	-	16.8 ± 0.3	403 ± 10	-
^{239}Pu	1008 ± 6	754 ± 9	254 ± 11	2.89 ± 0.05
^{240}Pu	281 ± 10	0.030 ± 0.045	281 ± 10	-
^{241}Pu	1370 ± 15	1030 ± 8	340 ± 16	3.03 ± 0.05
^{242}Pu	30 ± 2	< 0.2	30 ± 2	-

From the table, it is clear that both ^{239}Pu and ^{241}Pu can be fissioned by thermal neutrons. These isotopes may also be fissioned by resonance neutrons, and in that energy range, the cross-section values do vary dramatically in the region of a resonance.⁴²

For fast neutrons above about 1 MeV, all the plutonium isotopes are fissile. The cross sections for ^{239}Pu and ^{241}Pu decrease from the high thermal values, but level off to a value of about 2 barns. The fission cross sections for ^{240}Pu and ^{242}Pu rise abruptly in the range 0.4-1.0 MeV to values comparable to those of ^{239}Pu and ^{241}Pu . This "fast-fission" feature of plutonium, coupled with the thermal-fission aspect, makes this substance useful as the fuel in both thermal and power reactors.

D. Pyrophoric Properties

Throughout the past years, studies have been conducted on the ignition properties of plutonium.⁴³⁻⁴⁷ These were undertaken because the metal has proven to be quite pyrophoric under certain conditions.⁴⁸⁻⁵⁰ Spontaneous ignition of plutonium metals has resulted in severe fires in which great damage has occurred.⁵¹⁻⁵³ Plutonium fires can occur in air with massive forms of the metal, but are more likely to occur when the metal is in a more dispersed form.

The threat of fire arises mainly in these instances:⁴⁹

1. In work that involves turnings, chips, powders, or other finely divided forms of the metal.
2. In moist atmospheres, which allow unstable compounds such as plutonium hydride and hydrated plutonium oxide to form.
3. In processes forming certain alloys and metallic compounds that may be pyrophoric.
4. From impure residues, such as partially oxidized crucible skulls, which may ignite spontaneously.

Plutonium metal, in the massive and unalloyed form, oxidizes very slowly in dry air. From Table I, we see that the rate becomes rapid in a moist condition or when the metal is heated. Below the ignition temperature, the metal will not support combustion; above this temperature, oxidation occurs quickly. Factors that affect the ignition temperature include the rate of temperature rise, form of the metal, and previous exposure to oxygen.⁴⁶ In studies with small samples, temperatures in excess of 300°C have been reported. Recent data support a value of about 500°C for large samples of the metal.^{43,44}

In the form of the pure metal, plutonium is hard to ignite but when this does occur, it burns without flame to form inert PuO_2 in air.⁴³ A plutonium-metal fire is also difficult to maintain on a heat-conducting surface. The peak temperature reached during burning is directly related to the ratio of surface area to mass. Thus, when a given mass of metal is dispersed as turnings, these will reach a much higher peak temperature during burning. Table VI, prepared from the data of Felt,⁴³ contains some ignition features of plutonium metal.

TABLE VI Ignition Characteristics of Plutonium Metal

Material	Ignition Characteristics	Peak Temperature °C
Metal	Hard to ignite, burns without flame	825
Metal turnings	Instant ignition, rapid combustion	800-850
Skulls	Instant ignition, very rapid burning	620-635
Hood wastes (saw filings and drill turnings)	Becomes semimolten mass within seconds, white glow	1300-1350

Studies have shown that to completely prevent plutonium ignition and burning requires an inert atmosphere.^{43,45} Tests with argon-air, nitrogen-air, and carbon dioxide-air systems reveal that ignition does not occur for oxygen concentrations <5% unless finely divided plutonium is involved.⁴⁵

E. Properties of Transplutonium Nuclides

The element plutonium is the sixth member of a series of elements known as the "actinides."² The series extends up to element 103 and so includes many of the transplutonium elements. This series of atoms is so grouped because of the similar nature of their chemical properties. Much of our previous material in regard to plutonium can be applied to these elements with certain suitable modifications.

No known isotopes of the transplutonium group are stable. Most decay by alpha emission, although beta decay, electron capture, and spontaneous fission also occur.^{39,44} In general, the higher Z-number products have shorter half-lives, and this means greater decay rates. As the Z-number of the isotope increases, the amount or mass of the product that can be produced declines. For this reason, large quantities of these isotopes are not readily found.

The major differences between the transplutonic elements and ²³⁹Pu are higher decay rates, shorter spontaneous-fission half-lives, and more photons emitted per disintegration.^{30,39} Table VII, adapted from Denham,³⁹ compares some of these features with those of ²³⁹Pu.

TABLE VII Selected Transplutonium Nuclide Properties Relative to ²³⁹Pu

Isotope	Radioactive Half-life, years	Activity, dpm/g	Spontaneous Fission Half-life, years	Calculated Emission Rate, n/sec-g
²³⁹ Pu (α)	24,390	1.36×10^{11}	5.5×10^{15}	0.02
²⁴¹ Am (α)	453	7.61×10^{12}	2.0×10^{16}	0.62
²⁴³ Am (α)	7,370	4.44×10^{11}	-	-
²⁴⁴ Cm (α)	17.9	1.82×10^{16}	1.3×10^7	1.2×10^7
²⁵² Cf (α)	2.63	1.20×10^{15}	85	2.3×10^{12}
²⁵⁰ Bk (β ⁻)	0.85	3.74×10^{15}	6×10^6	2.7×10^5
²⁵⁴ Es (α)	0.76	4.12×10^{15}	7×10^5	2.9×10^5

III. HAZARDS OF PLUTONIUM

The hazards that are present in plutonium handling may be traced to some of the features discussed in Sec. II. These hazards arise from the radiation, toxic, nuclear, and ignition features of plutonium. The extent of these hazards depends upon many factors, among which are the form, amount, and isotopic composition of the plutonium and the type of operations performed.

A. Radiation Hazards

1. Relative Hazards

The relative hazards that radioactive substances present can be related to the nature of their emissions. For this reason, the nature of ionizing radiation was discussed in Sec. II.B.4. With regard to assessing the hazard, a major factor is the rate of energy loss in matter. In general, radiations with a high rate of energy loss are more effective in producing biological damage. On the other hand, radiations that quickly lose energy do not travel very far in any medium. By knowing the properties of the emission, one may refer to it as an internal or external hazard. If the emission has a short range in most materials, it will be of concern mainly as an internal hazard. If it has a long range in most substances, it will be significant as an external hazard.

a. Relative Hazard of Alpha Particles. The very short range of an alpha particle in any dense substance makes it of little concern as an external hazard. The most energetic alpha particle emitted by natural sources will just penetrate the outermost "dead" layer of skin. Thus, little living tissue will be affected. With the source on the skin surface, an alpha particle needs ~7.5 MeV to get through the dead layer. For most alpha emitters then, no living tissue will be damaged when the source is external to the body.

Once inside the body, the short range and high specific ionization (ion pairs produced per unit path length) of the alpha particle are cause for concern since it then becomes an internal source and may deposit its energy in localized living tissue. Any damage will be highly localized in a small volume about the deposition site. The more dense energy deposition along the alpha's path is known to be more effective in producing damage than that of less dense energy deposition from other radiations.⁵⁵ These factors make alpha radiation of vital concern as an internal hazard.

b. Relative Hazard of Beta Particles. Depending upon the particle energy, a beta or positron source may constitute an external hazard. Because of their greater range, beta particles of 70 keV are able to penetrate the dead layer of skin and hence deposit their energy in living tissue. Since most vital organs are at a depth of 5 cm or more, they will be unaffected by beta particles from sources having tissue ranges less than this, which is the case for most beta emitters. This source reduces an external beta source to primarily a skin-exposure problem. Since most beta particles are easily absorbed by a small amount of metal or plastic, the hazard will usually exist only very near the source.

An external hazard may result from X rays produced when a high-energy beta particle is stopped in an absorber (bremsstrahlung radiation). The amount of such X-ray production can be minimized by using low-Z-number absorbers. For positron emitters, the annihilation gamma rays require thicker absorbers to reduce the external hazard.

As an internal hazard, beta particles are not as significant as alpha particles. The greater tissue range of beta particles means that damage will not be as localized as for alpha particles. A much larger tissue volume about the deposited beta source will be involved. Some of the energy may actually be lost outside the small organ of concern. Annihilation gamma particles or bremsstrahlung may not even interact in the tissue mass, which means that some of the available energy will not be absorbed by the tissue. Also, the less dense energy deposition along the path of beta particles is less effective in producing damage than that for alpha particles. Nevertheless, damage may still be significant, so that beta emitters are of concern as an internal radiation source.

c. Relative Hazard of X and Gamma Rays. X- and gamma-ray sources are significant as external hazards. The great range of photons in air means the hazard may be significant at large distances from the source of radiation. To reduce the hazard, extensive absorbing matter (shielding) is often required. Also, scattered photons may contribute to the hazard. Direct-source radiation may be absorbed to ensure no hazard in one direction, while scattered photons may cause a hazard to exist in adjacent areas.

Because X and gamma rays do penetrate to such a high degree, the damage to tissue will extend throughout the body as photons pass through it. The deeper and more radiosensitive tissues will be exposed in an external gamma field. These tissues are more vital than skin, resulting in damage that affects the well-being of the body to a greater extent. This makes X and gamma rays of greater concern as external hazards than either alpha or beta particles.

From the standpoint of internal hazards, X and gamma rays are not as significant as alpha or beta particles. The longer tissue range of photons means less energy loss in a small tissue volume than for either alpha or beta particles. Since photon-energy loss occurs only at interaction sites, energy loss is not continuous along the photon path. In a small tissue mass, few interactions occur, since the path is small. For those that do occur, the density of energy deposition is similar to that for beta particles. Photons that do not interact in the organ carry away energy that is thus not deposited and therefore not effective in producing damage.

d. Relative Hazard of Neutrons. Neutron sources are significant as external hazards. Since the neutron moves rather freely through air and other matter, the neutron field may still be hazardous far from the source. Absorbing material can reduce the hazard, but the required amount is often

large. Scattered neutrons produced in the shielding material can also create concern in adjacent areas. In addition, radiative capture and inelastic scattering in the shield may result in enough gamma radiation to cause concern.

As neutrons travel through the body, tissue interactions extend throughout the body. The deeper, more radiosensitive tissues are exposed to the external neutron field. The amount of damage done is related to the neutron energy. Fast neutrons give up about 80-95% of their energy in elastic scattering processes with hydrogen in tissue. The recoil protons from these processes ionize atoms of the tissue. Intermediate and thermal neutrons lose their energy mainly in the absorption reactions: ${}^1\text{H}(n,\gamma){}^2\text{H}$ and ${}^{14}\text{N}(n,p){}^{14}\text{C}$.²⁶

Since neutron interactions produce recoil particles, protons, and gamma rays, the transfer of neutron energy to tissue will vary. The energy deposition from protons and recoil nuclei is more dense than that of electrons. This means that neutrons should be more effective in producing tissue damage than gamma rays. Depending upon the neutron energy, neutrons are 2-10 times more effective. On this basis, neutrons are more hazardous than gamma rays, although both are significant external hazards.

Neutron sources are not normally considered as an internal hazard. The lack of natural sources that emit neutrons and the physical properties of neutron sources make the chance of a neutron source being fixed in the body quite remote. This view could change if substances with high spontaneous fission rates become more readily available in larger quantities.

2. Biological Effects of Radiation

The harmful effects of radiation are due essentially to their ionizing effect on living tissues.⁵⁶ These tissues, composed of similar cells, perform special body functions. The cell, which is the basic unit of tissue structure, is composed of many elements, hydrogen, oxygen, carbon, and nitrogen being the main ones. Radiation passing through living cells ionizes or excites the atoms or molecules (groups of atoms) in the cell structure. This action may cause the molecule to break up into charged fragments called radicals and ions. These can produce further effects in the cell so that damage is caused in both a direct and an indirect manner. The total effect on cell processes will depend upon the type and amount of radiation absorbed. Cells will be affected in varying degrees up to cell death. If the extent of damage is large, tissue function may be impaired or even disabled.

a. Cell Mechanisms. The cell is usually composed of a nucleus surrounded by cytoplasm; both are encased in membranes. The nucleus is considered the control center of the cell. It initiates cell division (mitosis) and controls the repair of cell damage. The cytoplasm is a liquid substance, which is a functional participant in such cellular activities as absorption and excretion. Many types of body cells have a limited life span and divide at a certain stage of their life. This allows the daughter cell to take over the

parent-cell functions. As a cell divides, threadlike structures (chromosomes) appear in the nucleus. These contain the genes that determine hereditary characteristics. During cell division, the daughter cell receives a duplicate set of chromosomes and identical genes from the parent. If the process is normal, no alteration or changes occur in the chromosomes or genes. If change or damage does occur, mutation results that can affect either the daughter cell or future-generation cells. Some damage may be repaired by the action of the cell itself. If the cell is too badly injured, replacement can occur through mitosis of other healthy cells.

In man, cell types or lines are referred to as somatic or germ (gametes). The germ line gives rise to gametes or cells which can unite to reproduce the species. The somatic lines develop into the tissues of the individual. Since only the germ-line cells can be transmitted to future generations, damage to somatic cells is limited to the individual. Damage to the offspring of an individual may occur when there is damage to the cells of the germ line.

b. Radiosensitivity. All tissues of the body are susceptible to radiation damage. However, the cells that make up these tissues differ in appearance and function as well as in their response to radiation. For this reason, the radiosensitivity (response to radiation) of the body tissues also differs. To produce a specific biological effect, the necessary dose varies inversely as the relative sensitivity of the given tissue. Some factors that affect the response of tissue cells are: stage of development, metabolic activity, and division rate.⁵⁵ Bone marrow, lymphoid tissues, and the reproductive organs rank among the most radiosensitive. Muscle and bone mineral are among the least radiosensitive.⁵⁶

c. Relative Biological Effectiveness and Quality Factor. Although the types of damage produced by all radiations will be the same, the degree of response for an equal absorbed dose will differ, depending on the type of radiation, i.e., alpha versus gamma, etc. The absorbed dose D is defined as the energy imparted per unit mass of matter by ionizing radiation at the point of interest. The characteristic degree of response due to equal absorbed doses from different types of radiation (e.g., alpha versus gamma) is called the relative biological effectiveness (RBE). As used in experimental work, the RBE is often defined as the inverse ratio of the absorbed doses of two kinds of radiation that produced the same effect. To determine the RBE for a given type of radiation, one must define the exposure conditions as well as the effect studied. For this reason, the use of the RBE concept is generally restricted by convention to experimental radiobiology.⁵⁵

An important factor in arriving at the RBE of a given radiation is the linear energy transfer (LET). The LET expresses the rate of energy deposition measured along the track of an ionizing particle. X and gamma rays produce electrons that give low-LET tracks; alpha particles and neutrons, which produce protons and recoil particles, give high-LET tracks. It is generally believed that the more dense energy deposition from high-LET radiations is responsible for their usual greater effectiveness in producing damage.

In health-physics work, a factor is used to account for the variation in effectiveness to enable a comparison of the different radiation types in a mixed-radiation field. This quantity is called the quality factor (Q). It denotes that the effect of an absorbed dose is modified by the LET of the radiation. The Q is an assigned value; it is not arrived at by experiment. The factor applies to cases of normal radiation protection work. It is not applicable in the case of acute exposures at high dose rates, such as occur in accidents.⁵⁷ It provides a gross quality correction so that one may use a common scale for all types of radiation. The practical values of Q, suggested by the NCRP,⁵⁸ are shown in Table VIII.

TABLE VIII. Practical Quality Factors

Radiation Type	Rounded Q
X rays, gamma rays, electrons, or positrons	1
Neutrons, $E < 10$ keV	3
Neutrons, $E > 10$ keV	10
Protons	1-10 ^a
Alpha particles	1-20
Fission fragments, recoil nuclei	20

^aUse the higher value for round-off, or calculate by the methods of Ref. 58.

The common scale used to compare all types of radiation is called the dose equivalent (H). This may be defined as the product of an absorbed dose and other modifying factors. One of these factors is the quality factor. Another modifier, the distribution factor (DF), may be used to correct for nonuniform distribution in the case of internal sources. The rem is the unit used for dose equivalent and is numerically equal to the absorbed dose in rads times the appropriate modifying factors. The rad is a unit of absorbed dose equal to the absorption of 100 ergs of energy per gram of matter. The erg is a much larger energy unit than the MeV (by a factor of roughly one-half million).

d. Somatic Effects. Since damage to the somatic cells is limited to the individual, somatic effects include any types of damage that affect only the individual. A number of factors combine to make the effects differ for various conditions. The type of radiation is important, since some types are more effective in producing damage than others, as was discussed previously. The age of the individual is also a factor; children are more sensitive to radiation than adults. The absorbed dose is important because the total energy absorbed per gram of tissue can be related to the effects produced in the tissue. The time distribution of the exposure will also modify the effect; e.g., a "lethal dose" received over a relatively short period of time may not be lethal if protracted over a long time. The distribution of the dose is also important;

i.e., the effects are greater if the total body is irradiated rather than just a specific organ. In this case, a lethal whole body radiation dose may be far from lethal if it is limited to one specific organ.

Those effects that are observed may be loosely divided into early and late effects.⁵⁵ Early, or acute, effects occur at very high exposure levels and can be directly related to radiation as the cause. A broad picture of expected effects in man has been prepared by the NCRP⁵⁵ and is shown in Table IX.

TABLE IX. Representative Dose-Effect Relationships in Man for Whole-body Irradiation

Nature of Effect	Representative Absorbed Dose of Whole-body X or Gamma Radiation, rads
Minimal dose detectable by chromosome analysis or other specialized analyses, but not by hemogram	5-25
Minimal acute dose readily detectable in a specific individual (e.g., one who presents himself as a possible exposure case)	50-75
Minimal acute dose likely to produce vomiting in about 10% of people so exposed	75-125
Acute dose likely to produce transient disability and clear hematological changes in a majority of people so exposed	150-200
Median lethal dose for single short exposure	300

From the table it is seen that, without special techniques, it is difficult to find any immediate effects for acute doses of less than 50 rads. The absorbed doses shown are called midline absorbed doses. (An average absorbed dose of 1 rad of photons near the body midline would equal a dose about 1.5 times as high when measured in free air.) As dose increases, damage increases and, for large enough doses, death becomes more probable. In the dose range where survival may be the concern, the concept of the median lethal dose (LD_{50}) is used. This expresses the dose at which 50% of those exposed would die. As seen from the table, LD_{50} is estimated as 300 rads (about 450 rads free air dose). For partial body irradiation, much higher doses are required to bring about early or acute effects. These effects are rare, except following radiation therapy to localized portions of the body.⁵⁵

These effects, whether from partial or whole-body irradiation, are the result of unusual exposure conditions. That is, one would expect to encounter these exposure levels in an accident situation or single short incident, not under normal operations. Correct radiation-safety practice limits the exposure levels and duration so that none of these effects would be observed.

e. Late Effects. The problem in the study of late effects is that the elapsed time may be many years and may depend on the dose, making it

hard to relate the cause to the effect. Since the late effects may be caused by many other agents besides radiation, there can be no positive assignment of the cause in most cases. At best, it is known that radiation increases the incidence of these nonspecific injuries. The main effects are neoplasms (tumors), cataracts, life shortening, and effects on growth and development.⁵⁵

1. Hereditary Effects. Hereditary effects refer to those traits that can be transmitted from one generation to another in a given species. Inherited traits are determined by genes, and any mutation of a gene can result in an altered trait. Mutations occur spontaneously, and they also can be induced. Radiation (as well as other agents) produces the same effects, so that radiation only serves to increase the frequency of these effects. Even for high doses, increases in the rate are small.

Genes may be dominant or recessive. When a trait is determined by one of the genes, this is called the dominant gene. Mutations in dominant genes affect the offspring in the first generation. When genes are recessive, the usual result is some intermediate trait. Damage occurs only if the same altered gene is received from each parent. Unless the same type of mutation occurs in a large population, damage due to recessive genes will not show up readily for generations.

Some hereditary effects in man are caused by chromosome damage. These effects are early embryonic mortality, neonatal deaths, and certain diseases that are eliminated in the first generation.⁵⁵

Dominant mutations have a high probability of producing effects serious enough to terminate the line. On the other hand, recessive mutations have a small effect, but add to the number of mutations carried by the offspring. Added to this burden is the number of mutations caused by other agents. Thus, the total effect from the recessive genes may be greater than that of the dominant genes because recessives are eliminated less quickly. Recessive genes may not be eliminated for many generations.

3. External and Internal Exposure Aspects

The effects produced by radiation are the same, whether the exposure is internal or external, provided the absorbed dose and its distribution are the same.⁵⁵ The total organ dose is the sum of that from both types of exposure.

Radioactive matter may enter the body through inhalation, ingestion, absorption in wounds or punctures, and diffusion through skin. Once inside, these internal sources can end up in vital or critical organs. While they remain in the body, the irradiation is continuous. The amount in an organ decreases with time, due to radioactive decay and biological elimination. Each process has a half-life or time in which one-half of the initial amount decays or is eliminated, respectively. Both processes act to reduce the amount, but for long half-lives, the total elimination rate is slow. Thus, living tissue may be irradiated for a very long time.

Fig. 6. Mechanisms of Inhalation and Ingestion of Radioactive Matter

Consider the process of inhalation (see Fig. 6). Some of that taken in is immediately exhaled (out). Part of the inhaled matter is trapped by portions of the upper respiratory tract, and swallowed (A). This material is eliminated through the feces. During passage through the gastrointestinal tract, some of the material may be absorbed into the bloodstream (F_3) and transported to certain organs (F_2).

Some radioactive matter reaches the lungs and leads to direct lung irradiation while it is being retained (F_1). Some of this material may also be absorbed into the bloodstream and end up in certain organs (F_2). If the material is basically insoluble, part of the inhaled matter will be worked up from the lungs and swallowed (as in A). Some of the material will be retained in the lung and slowly taken up by body fluids.

Turning to ingestion, radioactive substances enter by eating, drinking, or by transfer of contamination from objects to the mouth. When such a substance is swallowed, the gastrointestinal tract (B in Fig. 6) receives direct irradiation during the time of passage. Again, if some of the material is soluble, part may be absorbed from the tract (F_3) and transferred to an organ (F_2). If insoluble, all will be eliminated.

The fractions F_1 , F_2 , and F_3 depend upon many variables, including the chemical properties of the material and its solubility in body fluids. In general, these fractions are not well known in humans. Data from animal experiments are used when applicable, but this information is not always available and sometimes not appropriate.

If radioactive material enters through open wounds or is transported through the skin, it may also pass to all portions of the body via the bloodstream. Little is known of the transfer fractions that apply in these instances. However, it is known that the solubility of the substance influences the transfer that occurs.⁵⁹ A larger fraction will translocate if the substance is soluble.

For internal exposure then, one must deal with continuous irradiation usually involving only limited parts of the body. In general, a substance deposits nonuniformly in tissue, leading to an uneven dose distribution. In terms of the potential for producing biological damage, short-range, high-LET radiation is the most important. Because of the fixation of the material in the

body, internal exposure cannot be readily controlled. It is therefore necessary to direct a conscious effort toward eliminating any intake.

In the case of external sources, exposure can be more readily controlled. One can limit the time of exposure and reduce the magnitude of the radiation field through distance and/or shielding. Source intensity and, hence, exposure rate decrease rapidly with distance from the source. When the source dimensions are quite small, this decrease varies inversely as the square of the distance. A shield is a dense substance which reduces the radiation field for those cases in which the worker must be close to the source. The dense substance provides a large amount of closely packed matter, which increases the number of radiation interactions. This increase tends to remove radiation more quickly and hence results in less intensity. Any one or all of these three factors may be used in the control of external exposure.

On the other hand, certain external sources can deliver a very high dose in a very short time. Often the whole body, not just part of it is exposed. These features are usually absent in internal exposure, but require attention in the case of external sources.

Accelerators, X-ray devices, and reactors require special care in their use to reduce the chance of external exposure. When charged particles such as electrons and protons are accelerated, they become more hazardous as they gain energy. This is due to the increased penetrating power and the nuclear reactions they are able to cause at high energies. Reactors are a potent source of both neutrons and gamma rays. Moreover, samples placed in the reactor become radioactive and require special care in handling during removal. The spent fuel removed from a reactor will contain gross amounts of fission products which will be highly radioactive. A serious whole body dose can be received in a very short time from any of the above sources.

4. Radiation-protection Standards

The task of setting safe radiation limits is a vital and yet, very difficult, undertaking. It is vital because workers must be protected from the harmful effects of ionizing radiation. It is difficult because so many factors enter into the effects that radiation produces. Though considerable data have been gathered and studied, many complex areas need more work before firm conclusions can be reached.⁵⁵

Since some unknowns still exist, the setting of limits involves judgments that cannot be wholly based on scientific knowledge. This process depends upon the concept of an "acceptable risk." That is, the benefits are weighed against the potential harm, and limits are set at some level at which the most benefit to mankind will accrue. From time to time then, limits are revised as new knowledge is gained. This reflects an attempt to replace assumption by fact in order to arrive at a better value judgment.

The present radiation-protection standards for internal and external exposure that govern ERDA Contractor operations⁶⁰ are shown in Table X.

TABLE X. Radiation-protection Standards for External and Internal Exposure for Individuals in Controlled Areas

Type of Exposure	Exposure Period	Dose or Dose Commitment, rem
Whole body, head and trunk, gonads, lens of the eye, red bone marrow, active blood-forming organs	Year	5
	Calendar quarter	3
Unlimited areas of the skin (except hands and forearms). Other organs, tissues, and organ systems (except bone).	Year	15
	Calendar quarter	5
Bone	Year	30
	Calendar quarter	10
Forearms	Year	30
	Calendar quarter	10
Hands and feet	Year	75
	Calendar quarter	25

The standards shown in the table set the limits that should not be exceeded. However, the goal is to keep all exposure as low as reasonably achievable; thus, operational guides may need to be imposed. These guides are used as signals when the exposure level approaches or reaches a certain value. This permits an awareness that control may be breaking down before the limits are exceeded. Also, the guides and standards serve as yardsticks for evaluating the relative hazards of a radiation field. That is, if no controls are imposed, a certain dose rate from a given source may deliver a dose in excess of the limits. Depending upon what this excess is, the relative hazard and the necessary controls will vary. For this reason, a measure of the radiation field from a given source is needed for the purpose of evaluating the radiation hazard.

5. External Dose Rates from Plutonium

The external dose rate from plutonium includes contributions from X rays, gamma rays, and neutrons. Electrons (beta particles) have been ignored since they are of low energy and will usually be absorbed by the gloves of the handling enclosure. The actual dose rate from a sample of high-exposure recycled plutonium will be a function of the isotopic composition. From Fig. 4, it can be seen that such a sample will contain a fairly large total fraction of ^{238}Pu , ^{240}Pu , ^{241}Pu , and ^{242}Pu . Also, the daughter products ^{237}U and ^{241}Am contribute to the gamma dose rate as these products build up. The net effect of the isotope content is to increase the expected dose rates as the total fraction present in the sample increases.^{61,62} The calculations of Roesch,³¹ Birchall,³² and Arnold⁴¹ have provided a basis for estimating external dose rates from

plutonium. These results have been summarized^{29,38} and have been adapted for use in this section. The advent of high-exposure plutonium has greatly increased the external radiation hazard, especially in regard to extremity exposures. Not treated here is the impact of fission-product contamination on the external dose rates from recycled plutonium.

a. Surface X and Gamma Dose Rates. X rays from plutonium occur as the result of the internal conversion process. This process is an alternative mode of gamma decay. An excited nucleus that may emit a gamma ray transfers this energy instead to one of the outer electrons of the atom. This ejects the electron, and this loss leaves the atom in an excited state. When the atom returns to the ground state, a discrete-energy X ray (fluorescent X ray) is emitted. Fluorescent X rays also result from gamma absorption in high-Z-number substances and these add to the X-ray total.

Gamma rays from alpha decay, spontaneous fission, and the fission products of a fissioned atom are sources that increase the gamma contribution. In addition, X and gamma rays are emitted by ²³⁷U and ²⁴¹Am, which will increase the total dose rate as time goes by.

The surface dose rate can be divided roughly into two portions, that from photons below about 40 keV and that from those above. Table XI gives the surface dose rate through a layer of about 50 mg/cm² of tissue equivalent material from massive plutonium metal.³¹

TABLE XI. Photon Surface Dose Rates^{31,38}

Isotope	Surface Dose Rate, rad/hr	
	<40 keV	>40 keV
²³⁸ Pu	980	2
²³⁹ Pu	0.61	0.056
²⁴⁰ Pu	14	0.62
²⁴² Pu	0.19	0.11
²⁴¹ Am	0.09t ^a	0.10t
²³⁷ U	0.56(1 - e ^{-0.102t}) ^b	23(1 - e ^{-0.102t})

^aTime (t) in days since plutonium was purified; valid for times <<14 yr.

^bTime (t) in days since plutonium was purified; valid for times <<14 yr.

From Table XI, the total dose rate from X and gamma rays at the surface becomes

$$D \text{ (rad/hr)} = 982f_{238} + 0.67f_{239} + 14.6f_{240} + 0.3f_{242} + 0.19f_{241}t + 24f_{241}(1 - e^{-0.102t}),$$

in which the symbol f_A stands for the fraction by weight of the isotope of mass number A . By only using the dose rates for >40 keV, we can obtain an expression for the gamma or "hard" portion of the surface dose rate. As fuel exposure increases the isotopic composition, the photon surface dose rates from the recycled fuel also increase.

b. Neutron Dose Equivalent Rates at Emitter Surface. Neutrons arise from two sources: spontaneous fission and (α, n) reactions in light elements. Both of these processes produce fast neutrons. For the fission neutrons, the mean energy is about 2 MeV,⁴² and for (α, n) reactions the mean energy is estimated as between 0.75-1.5 MeV.³⁰ If we assume a mean energy in the range 1-2 MeV, the dose equivalent rate would be 1.28×10^{-4} rem/hr per neutron/cm²-sec.²⁶ Roesch³¹ has derived the expression for the neutron flux density at the surface of a metal sphere. With his result, and the above conversion factor, the dose equivalent rate at the sphere surface is

$$H_N \text{ (rem/hr)} = 2.82 \times 10^{-4} (\eta/\rho) M^{1/3},$$

in which η/ρ is the number of neutrons emitted per gram per second and M is the mass of plutonium in grams. Using the data from Table III for η/ρ , the expression becomes

$$H_N \text{ (rem/hr)} = (0.65f_{238} + 0.26f_{240} + 0.5f_{242})M^{1/3}$$

for the neutron component from spontaneous fission. For the (α, n) reactions,

$$H_N \text{ (rem/hr)} = (609f_{238} + 0.12f_{239} + 4.5f_{240} + 0.05f_{242})M^{1/3} \text{ for PuF}_4$$

and

$$H_N \text{ (rem/hr)} = (3.95f_{238} + 0.01f_{239} + 0.05f_{240})M^{1/3} \text{ for PuO}_2.$$

From the two expressions above, it is seen that the ^{238}Pu isotope greatly affects the total neutron emission. Estimates of the $^{238}\text{Pu}(\alpha, n)$ neutron activities for other light-element compounds have been derived.³⁴ The neutron contribution from a plutonium sample does not change with time as does the photon portion. However, in light element compounds, the ingrowth of ^{241}Am will increase the (α, n) output.

c. Other Dose Rates. Not only may the surface dose rate be of concern, but also that at a given distance from the source. According to Roesch,³¹ for photons, the dose rate at some distance r is related to the surface dose D_s . The expression involves the projected area A of the plutonium sample and is given by

$$D \text{ (rad/hr)} = \frac{D_s A}{2\pi r^2},$$

where D is the absorbed dose rate at some distance r (cm) from the surface of the sample.

For neutrons, the estimate of the dose equivalent rate, at some distance r (cm), may be obtained from

$$H \text{ (rem/hr)} = (0.024f_{238} + 0.010f_{240} + 0.018f_{242}) \frac{M}{r^2},$$

in which H is the spontaneous neutron dose equivalent rate. This should be valid when the distance r is roughly six times the largest sample dimension.

B. Toxic Hazards

1. Toxicity of Internal Emitters

The toxic aspects of an internal source are governed by a number of factors. Among these are the type and energy of the emitted radiation, since these features alter the biological effectiveness of a given organ dose. The physical and chemical forms enter in because these influence the uptake and distribution patterns in the body. Even the age and species of the subject, as well as the mode of entry, modify the toxic effect.

One problem is to pick out the organ that is most affected by the given radioisotope. In many cases, a substance will be taken up by a number of body organs. The concept "critical organ," or that organ of the body whose damage by the radiation results in the greatest damage to the body, is used to deal with this problem. A number of factors affect the choice of a critical organ. Some organs are more essential to the body than are others, and slight damage to these may affect the body more than heavy damage to some others. Other factors, such as the radiosensitivity of the organ and the fraction of the radioisotope accumulated in the organ, are also important. When little uptake by the body occurs, the greatest harm may be produced in the organ of entry.

From the standpoint of absorption into the body, the solubility of the substance in body fluids plays a major role. It is not so easy to tell whether a given substance will be soluble in body fluids. However, as a mode of entry, ingestion is of concern only for the soluble form of the isotopes. In the case of elements not normally required by the body, uptake by ingestion is minimal.

When uptake occurs, one is concerned with the transfer of the material to some organ. Only a fraction of the ingested matter will go into the bloodstream. Of this, only a fraction will go into the organ of concern (see Fig. 6). To add to the problem, almost equal portions of the substance may end up in a large number of organs. This increases the problem of choosing "the" critical organ, since several organs may be essential to proper body function.

For the problem of inhalation, both soluble and insoluble matter must be treated. Other factors are particle size and density, as well as the

physiology of respiration.^{63,64} The movement and retention of matter in the respiratory tract are sensitive functions of the particle size.⁶⁵ In general, for size in the range 5-10 μm , these particles are trapped and easily removed from the upper respiratory tract. For sizes below this, the chance of the particle reaching the deep portions of the lung increases. The mechanism is quite complex so that estimates of the transfer of matter by this mode of entry are poor.

One must also treat the matter of elimination. Even though the substance is deposited in an organ, the time of stay in the organ is not indefinite. A fraction of the substance is eliminated from the organ as time passes. For a radioactive substance, a certain fraction decays per unit time, but some of the material leaves the organ because of biological processes. One must have knowledge of the net growth in a certain organ to assess the effect of the radioisotope deposit. The ICRP⁶⁶ assumes that each organ can be treated as a separate compartment in which a substance is eliminated at a constant rate. This means that each organ has a half-life for biological elimination of a given substance. This model provides a simple means of obtaining a rough estimate of the retention in an organ.

Given the amount of a radioisotope in an organ, one can then estimate the dose equivalent (H) rate. If one has a standard for the allowed H rate, then one can arrive at a permissible amount or organ burden. This will be the amount of the radionuclide in the organ that delivers the allowed H rate.

A much greater problem is to estimate the amount of a substance in an organ that results from intake into the body. Given certain intake rates, one can estimate values for the permitted concentration in air and water for the radioactive materials. This concentration would not result in an accumulation greater than the organ burden for an assumed daily intake. Of course, to make this estimation requires the use of many of the above-mentioned factors. At any rate, the ICRP⁶⁶ has made recommendations in regard to concentration values (MPC) and organ burdens (MPBB). The MPC (maximum permissible concentration) gives the allowed concentration in air or in water for a given radioisotope. Intake at the MPC rate for 50 yr will not result in an organ accumulation greater than the organ burden. The term maximum permissible body burden (MPBB) denotes the amount of the radioisotope in the total body such that the critical organ burden is not exceeded.

2. Toxic Features of Plutonium

There are many reasons why plutonium ranks among the more toxic radioactive materials once absorbed in the body. The alpha particles emitted by this substance have relatively high energy (see Table II). Not only is their range in air short (~3.8 cm), but the tissue range is extremely short (35-40 μm). The dense, highly localized energy loss from the plutonium alphas means a greater biological effectiveness in tissue. Once inside the body, the beta and electron contributions also take on added importance.

Soluble plutonium enters the body fluids and is transported throughout the body, ending up in liver and bone. Bone is presently taken as the critical organ in the case of soluble plutonium.⁶⁶ Plutonium is found both on the bone surfaces and in the bone-marrow spaces.⁶⁷ The uneven, initial surface deposits result in high alpha exposure to the sensitive, active bone-lining cells. Bone growth and other processes remove the uneven surface plutonium and re-deposit it at other sites or bury it under new bone.⁶⁸ During these processes, an uptake of plutonium in the marrow occurs.⁶⁷ In man, plutonium is tightly bound in the bone so that the excretion rate is slow. In the past, the biological half-life has been taken as 200 yr,⁶⁶ but recently the ICRP has suggested a new value of 100 yr.⁵⁹ Since the radioactive half-life is also long for ²³⁹Pu, only a small portion is removed during a life span.

The body burden for ²³⁹Pu with bone as the critical organ is only 0.65 μ g. This means that only a very small amount of ²³⁹Pu need get into the body to give a significant deposit. This also increases the risk, since high dose rates may result from relatively small intakes, in terms of the mass of material.

The lung is the organ of interest when one considers insoluble plutonium. Much of the initial portion taken into the lung is cleared rapidly. That which is retained after about the first day is in the deep regions (pulmonary) of the lung.

Some of the insoluble material will be slowly worked back up the throat and swallowed (see Fig. 6). The remainder is cleared by movement into the lymph nodes.⁶⁴ If the intake is soluble, the material will be taken up by blood and deposited in bone and liver. Deposition of matter in the lung is seldom uniform.⁶⁹

In the past, the half-life of insoluble plutonium in the lung was taken as one year,⁶⁶ but recent evidence suggests that 500 days or more may be more appropriate.⁵⁹ In terms of mass, the lung burden is 0.26 μ g (0.016 μ Ci). Even a small intake of insoluble material then represents an important quantity with respect to the lung burden. Also, material that moves to the lymph nodes tends to accumulate there. This long-term retention may require further evaluation of lymph-node damage, but presently the lung remains as the critical organ.⁵⁹

Most human exposure to actinide elements has occurred either from inhalation or absorption through wounds. Animal experiments have yielded estimates of absorption from lung to the critical organ of 1% to 10-15%. In the case of wounds, some remains at the site of entry, some is transported, if soluble, to the skeletal system, and some may migrate to the lymphatic system. For wounds, deposition is assessed through the use of instruments that measure the intake amount at the site. However, no model has yet been worked out to relate the amount at the site to that which is eventually deposited elsewhere.

Ingestion is not considered a major mode of entry, since absorption through the gastrointestinal tract is very low.^{63,70} Diffusion through intact skin is negligible,⁶³ whereas injection is not a very likely mode of entry. However, plutonium in organic solvents or acids may enter the body when in contact with skin. Regardless of the mode of entry, though, plutonium is rapidly cleared from the bloodstream and fixed in tissues.

3. Toxic Effects of Plutonium

The toxic effects of plutonium in humans must be deduced from animal studies. In large animals, with life spans of several years, lung cancers, tumors of the lung capillaries, bone tumors, nasal tumors, and liver tumors have been observed.^{68,71,72} These late effects in general are produced by large doses.

Lung cancer in dogs has been noted at levels about 30 times the present lung-burden value for man.⁷³ Bone cancer occurs in dogs for levels about 25 times the present bone burden.⁶⁷ No leukemias were reported in dog studies, but have been observed in rats. The high levels of exposure used in these studies may produce a spectrum of cancers that would differ from that found at low levels.⁷¹ For this reason, present long-term dog experiments are underway at low levels. These levels are in the range from 1 to 30 times the present body burden. In small animals, changes in the lungs were observed for low and high doses; this was attributed to uneven distribution of plutonium.^{72,74}

Langham⁶³ has discussed acute effects of plutonium. The basic mechanism seems to be a breakdown of the blood-forming system. These effects were observed only when massive amounts of plutonium were given to the animals. No data exist for man, but it is estimated that to observe acute effects would require an ingestion of pounds of plutonium or the inhalation of gram amounts.

For late or delayed effects, studies that compare radium to plutonium toxicity have been made. Like plutonium, radium also concentrates in bone. Because of its early use in dial-painting industry and in medicine, human exposure data are available for direct study.⁷⁵ The data on the delayed toxicity of ²²⁶Ra in man has allowed a direct evaluation of the body burden for radium. For this reason, radium is used as the standard for bone seekers. The results of the comparative studies indicate that plutonium is 5 to 10 times as hazardous as radium on the basis of the absorbed dose in bone.⁷⁶

Turning to human data on plutonium effects, no specific injuries have been noted.⁷⁷ The largest group studied has been 25 workers exposed during 1944-1945. A recent follow-up study showed estimated bone burdens of 0.1-10 times the currently used level.⁷⁸ Although no injuries have yet appeared, the long latent period associated with some late effects of radiation makes it prudent that such studies continue.

4. Toxic Index

One may use the body-burden value as a toxic index to arrive at the relative hazard of a given amount of material. That is, the relative hazard is assessed by comparing the given amount of the substance to its body-burden value. Table XII contains these values for selected nuclides with bone as the critical organ. For the microcurie values ($1 \mu\text{Ci} = 2.22 \times 10^6 \text{ dpm}$), the relative hazard varies little. However, using microgram values, the relative hazard may differ by several orders of magnitude. This points out the difficulty in control, since the release of the same mass of each nuclide gives vastly different activities. The table also shows the ICRP concentration values (MPC). These are used to assess the relative hazard of measured airborne plutonium concentrations.

TABLE XII. Body Burdens and Maximum Permissible Concentrations⁶⁶

Radioisotope	Body Burden		MPC in Air, ^a $\mu\text{Ci}/\text{cm}^3$
	μg	μCi	
²³⁸ Pu	2.3×10^{-3}	0.04	2×10^{-12}
²³⁹ Pu	0.65	0.04	2×10^{-12}
²⁴⁰ Pu	0.18	0.04	2×10^{-12}
²⁴¹ Pu	8.0×10^{-3}	0.9	9×10^{-11}
²⁴² Pu	13	0.05	2×10^{-12}
²⁴¹ Am	1.5×10^{-2}	0.05	6×10^{-12}

^a These values are similar to the Concentration Guides (CG) of Ref. 60.

C. Criticality Hazards

The mere presence of any fissile material does not mean that a criticality hazard will exist. For such an event to occur, given the proper setting, a certain amount of the substance must be present. As discussed in Sec. II.C.2, this amount is called the critical mass. This term should not be taken to mean that only the mass of the fissile substance is a factor. Indeed, many other factors such as the shape, form (solid or solution), nature of the surroundings, and neutron absorbing features of the system enter in. Given a favorable set of parameters, there is then a minimum critical mass, which does refer to the total amount of the fissile matter. If this value is not exceeded, no criticality will occur. Table XIII presents some minimum critical values for the isotope ²³⁹Pu and points out the influence of some of the factors. Note that the presence of other isotopes in the sample will modify the values obtained for ²³⁹Pu alone. The values in the table under the column marked "recommended value" include safety factors. Use of these values will provide an extra measure of safety in plutonium-handling operations.

TABLE XIII. Basic Nuclear Parameters for ^{239}Pu (Ref. 79)

	Recommended Values	Minimum Critical Values
Mass, kg		
Solution	0.22	0.51
Metal	2.6	5.6 alpha phase
	3.5	7.6 delta phase
Diameter of infinite cylinder, in.		
Solution	4.2	4.9
Metal	1.4	1.7 alpha phase
	1.8	2.1 delta phase
Thickness of infinite slab, in.		
Solution	0.9	1.3
Metal	0.18	0.24 alpha phase
	0.22	0.28 delta phase
Solution volume, liters	3.4	4.5
Chemical concentration of aqueous solution, grams of isotope per liter	6.9	7.8

Despite care in handling and safety in design, an unwanted criticality can result. The incident occurs with no warning, so that there is no time to invoke safety measures. That is, the system is subcritical one instant and then, suddenly, critical. When one of these accidents begins, nothing stops the process until the system returns to the subcritical state. In most cases, a single short burst of radiation occurs in a fraction of a second and the process is not sustained. In other cases, a sustained self-controlled reaction for a rather indefinite time period may result. Both types have occurred on occasion, but the more likely event is a burst type.

In the burst type of incident, these effects can result: (1) high radiation levels, both n and gamma, which can be lethal for a considerable distance; (2) the release of enough heat to melt metals in some parts of a contained system; and (3) contamination of the nearby area severe enough to present a hazard to personnel for perhaps months;⁸⁰ and (4) high gamma residual radiation levels from fission products.

Such a burst-type event may occur in an unshielded system of low initial radioactive content. It may also occur in highly radioactive systems such as reactors and processing plants.⁸¹ In the latter case, the presence of large amounts of shielding would greatly reduce the initial radiation exposure. However, the release of airborne radioactive material would result in extensive contamination. In the case of the former, the contamination would be high only in the local area of the event. The minimum value for the burst size in a system may be estimated as 10^{15} fissions for alarm purposes, 10^{16} fissions for dosimetry applications.⁸² For a water-moderated assembly, one may expect 10^{17} - 10^{18} fissions in a short-burst incident. For metal assemblies, accidental bursts of 10^{17} - 10^{19} fissions are assumed possible.⁸¹

From the data on the mockup experiment of the Y-12 incident,^{83,84} it is possible to compute the prompt neutron and gamma doses as a function of distance from a burst of 10^{18} fissions.

Fig. 7. Prompt Radiation Dose from a Burst of 10^{18} Fissions. Total absorbed dose is ≥ 600 rads up to 20 ft. ≥ 100 rads up to about 50 ft.

These are shown in Fig. 7. From the figure, one can obtain the total dose at some distance and scale this up or down for an assumed number of fissions. No shielding effects have been taken into account nor changes in the n/γ ratio for other types of critical assemblies. The data may be used to obtain a rough estimate of the doses possible in an incident involving a moderated system.

To estimate potential doses from an incident in a metal system, the data from the Health Physics Research Reactor burst operations may be used.⁸⁵ The n/γ ratio of this system varies, but is in the range of 5-9. On the basis of data taken for $n/\gamma = 5.7$, a burst of 10^{18} fissions in a metal system would produce 4000 rad_n and 700 rad_γ at 10 ft. Assuming no shielding, this would yield total doses of ≥ 600 rads up to about 30 ft and ≥ 100 rads up to around 70 ft.

Note that these estimates are tied to certain n/γ ratios. These ratios may vary from system to system, as well as in a series of bursts using the same system. In general, for a moderated system, one would expect the gamma dose to be the higher; in a metal system, however, the neutron dose would be expected to be the higher one.

For a burst of about 10^{18} fissions, dispersion of the fissile substance and the fission products would follow. Heavy local contamination would result, giving a very high residual dose rate. This dose rate decreases very rapidly shortly after the burst, but may still be on the order of tens of rad/hr one hour after the event.⁸¹ In addition, the dispersal of the plutonium would mean very high alpha contamination levels.

The criticality incidents that have occurred in AEC operations are described briefly in Ref. 86.

D. Fire and Explosion Hazards

As in the case of a criticality incident, the hazards from a fire or explosion exist only if the event occurs. For this reason, the emphasis is on

prevention of the incident. Should the incident occur, the potential hazards would be personnel injury or death, property damage, and loss of equipment.

The components necessary to initiate and sustain a fire are: fuel, heat, oxygen, and a chemical chain reaction.⁸⁷ That is, a substance (fuel), raised to the proper ignition temperature (heat) in the presence of air (oxygen), will ignite and continue to burn (chain reaction). So, to prevent the start of a fire (or to put out one already started), one must:

1. Remove one of the three elements: fuel, heat, oxygen.
2. Inhibit the chain reaction.

With respect to fire prevention, these efforts are directed at achieving the first goal. One may inert the system (remove oxygen). If this is not practical, one may center efforts toward removal of fuel sources or heat (ignition) sources. That is, analyze the process for conditions that allow fire to start and attempt to remove these. If one removes all fuel sources, or all ignition sources, the fire-prevention goal will be realized. However, sometimes the process itself and the type of enclosure used preclude removal of all fuel or ignition sources.

Many of the sources of fire and explosion have been treated in the literature.^{5,7,11,44,88} In Sec. II.D, we discussed the features of plutonium that make it a source of ignition at times. Among the other items that can be classed as ignition sources are chemical reactions, services and equipment that may produce heat or sparks, and excessive heat from alpha and fissile material.^{5,11,34} Explosion sources include flammable gases and solvents, finely divided metallic dusts, compressed fluids, equipment, and chemical reactions.^{5,11,88}

In addition to the ignition sources above, many fire hazards can be found within the enclosure. In the case of gloveboxes, the enclosure itself may be constructed of many combustible substances.¹³ Such items as the gloves, exhaust filters, plastic panels, and vinyl bags may readily burn in a fire from some other source or even support combustion.⁴⁴ Fires arising from ignition of oils, solvents, rags, and paper can lead to rapid heating in the system.⁸⁹ Although the initial source of ignition may be small, the fires that arise in the secondary materials can be very serious and difficult to control.

A fire or explosion in the enclosure may cause either a gradual or a sudden pressure increase, which can cause failure of the enclosed system. Based upon the speed of the ignition (or flame) propagation and the rate of energy release, combustion explosions fall into two types.⁸⁸ The usual type occurs because of the increased pressure from a thermal expansion caused by the propagation of ignition through the combustible mixture. This movement of the flame occurs at less than the speed of sound. The peak pressure that is reached is low, seldom above 130 psi (pounds per square inch). Moreover, this pressure can be reduced to only a few psi with pressure-relief devices.⁸⁸

The other type is an extremely fast explosion. The flame advances at speeds much greater than the speed of sound, and the peak pressure may reach many thousand psi. This type is called a detonation and cannot be effectively vented.⁸⁸

The ability of the enclosure to withstand the increased pressure depends upon the duration, rate, and extent of the pressure rise. In general, most combustion explosions start out as the first type, and a few are of the second type. This is fortunate, since in the case of a weak structure, such as a glovebox, the enclosure would not be able to contain a detonation. Smaller gloveboxes are more vulnerable than larger ones, since more of the volume may be involved in the explosion, and this increases the pressure load.⁸⁸

Another type of explosion hazard of concern involves those that arise in experimental equipment and are rather anomalous in regard to pattern. These serve to point out that not every case of overpressure results from combustion processes. These types not only produce an overpressure, they also create missile hazards if the device or setup shatters into many pieces. Of course, many explosions that breach the containment also produce missiles in the process.

In the case of plutonium fires or explosions, a number of hazards may arise.⁷ These are briefly discussed in the following sections.

1. Personnel Hazards

If the containment is breached, plutonium will be dispersed into the area. This presents an immediate hazard in regard to inhalation and a subsequent hazard due to widespread contamination. Because of the extreme toxicity of plutonium, the extent of the hazard may be severe, even if only a small amount of matter is involved. In addition, severe injuries due to the blast pressure or flying missiles may occur in explosion. Added to this, wounds caused by contaminated objects could result in internal plutonium contamination.

2. Contamination Hazards

A fire or explosion that results in the release of plutonium may contaminate large areas of a plant. This would require expensive and extensive decontamination.⁷ Present information suggests that, outside of the facility itself, little plutonium will be dispersed beyond the plant boundaries.⁹⁰⁻⁹³ This is attributed to two factors: Burning plutonium produces large particles that tend to settle out quickly, and airborne plutonium particles become readily attached to surfaces.⁹¹

3. Plant Damage

The damage to the plant from a fire or explosion can be quite extensive.⁵¹⁻⁵³ This damage not only includes the physical damage, but also that caused by the loss of items too badly contaminated to reclaim. To this must

be added costly delays in vital programs. These arise out of the inability to replace crucial, damaged equipment and to use the area in which the incident occurred. Other areas may also be affected either by the event directly or by tracking of contamination following the accident.

4. Criticality Hazards

A critical mass may be formed during an incident when a metal shape is changed by dispersal or by melting. The process of fighting a fire may introduce moderators, such as water, which could enhance the chance of a criticality. An explosion could cause otherwise safe amounts to be blown together, thus creating a criticality. The hazards produced by this type of event are those discussed in Sec. III.C.

E. Hazards of Transplutonium Nuclides

In general, the same hazards are present in handling transplutonic nuclides as in handling ^{239}Pu , except for degree. Because of higher decay rates in most cases, these substances are more hazardous on a mass basis.^{93,94} Hand exposures could be quite high and may be the limiting dose factor for glovebox-handling operations. Neutron dose rates will be greater because of the shorter spontaneous fission half-lives (see Table VII). The self-heating and spontaneous-ignition effects in these substances are much greater than in ^{239}Pu , and foils may even ignite in air.³⁹ The threat of criticality is less than for ^{239}Pu because sufficient amounts of these substances have not been produced. However, they are fissioned mainly by fast neutrons, and the critical mass needed may be less than that for ^{239}Pu . The biological patterns have been studied for some of the transplutonic elements, but no firm estimates of any differences in damage were made.^{95,96} In total, these factors combine to indicate that, as a group, the transplutonium nuclides may be more hazardous than ^{239}Pu .

IV. CONTROL PROCEDURES

A. Introduction

The type and extent of control measures used in plutonium work will vary. Many factors enter in, and only some of these were briefly mentioned in Sec. I.B. In some cases, plutonium is handled in open-faced hoods, but in most instances, it is handled in semiremote units (gloveboxes) or remote enclosures (hot cells).^{6,7,11,18,89,97} The features of a particular handling operation may dictate special control measures, as does the state of the plutonium being used. Low levels ($<1 \mu\text{Ci}$) of nondusty plutonium samples do not require the degree of control that higher levels do. Sealed or clad plutonium, regardless of amount, may be handled in hoods rather than gloveboxes for certain steps in the process. Several reports deal with both the hazards and control measures of certain programs.^{34,54,61,64,98-102}

The laboratory handling of plutonium differs from that found in a plant or production facility.⁷ In general, on the order of 10 mCi or less of plutonium is handled in open hoods; gloveboxes are used for larger amounts. In plants in which gram or kilogram amounts are used, gloveboxes are used almost exclusively. For dusty operations or for plutonium in other pyrophoric forms, these call for gloveboxes with inert atmospheres. In processes involving irradiated plutonium samples that have not been separated from fission products, hot cells are used to provide shielding. In the future, glovebox handling of plutonium may be precluded.

In all these systems, the enclosure forms the primary confinement of the material. Added to this, the airflow design and filter placement, as discussed in Sec. I.A, will contain any releases. The module, room, or building itself will also add a degree of containment.

Transfer of items into and out of the enclosure must be done so as to preserve the barrier integrity. For a closed system, such as a glovebox, the design often includes plastic-bag ports or sphincter valves which allow such transfers.^{5,6} Plans for changing gloves as well as windows must also be thought up for glovebox procedures.

The probability of accidents such as spills, unexpected chemical reactions, small fires, or explosions should be considered.⁵ Safety design will attempt to overcome or minimize the effects of such incidents. However, since these events may still occur, it is extremely important that personnel be trained in methods of combating such episodes. Needless to say, plans must be formed that treat the problems arising out of such accidents.

Regardless of the design excellence, the worker must also follow certain approved, albeit regimented, practices, which should be spelled out in written work instructions. These measures should include such things as:

1. Remove all unnecessary equipment from the enclosure before starting a new project.
2. Use absorbent matter or strippable paint on the surface to avoid contaminating the floor of the enclosure.
3. Limit or avoid the use of pointed tools, wire, or sharp objects, and quickly report any punctures, wounds, or scratches obtained while performing plutonium work.
4. Follow good housekeeping practices: Clean interior surfaces often, clean spilled matter at once, remove solid waste promptly, limit amount of solvents and other combustible material, and segregate pyrophoric matter.⁵ Do not let dust pile up!
5. Inspect gloves often for signs of rupture, wear, pin holes or deterioration. Be aware that acids and other mixtures attack the gloves. Also, gloves fail rapidly in work with oxides of high specific alpha activity.³⁴ Change gloves frequently to avoid mishaps (preventive measure).
6. Remember that stored vessels that are not vented can result in pressure buildup leading to rupture and spillage. Also, the storage of high-specific-activity alpha emitters in solution may result in heat evaporation of the liquid. This process may occur in conjunction with breakdown of the liquid (radiolysis) in the solution, causing gas buildup. For a very high specific activity, such as for ^{238}Pu , gas bubbles rising to the surface may burst and form a very fine oxide dust, which can grossly contaminate the enclosed system.³⁴
7. Avoid splashing of liquids or spraying them during enclosure operations, and keep liquid containers in a secondary container.
8. Use latex or surgeon's gloves in addition to those that are part of the enclosure.
9. Do not perform plutonium work with open wounds or scratches unless you have obtained prior approval from medical authority.
10. Know what your responsibilities are in regard to work performance and safety procedures.

The above listing is not meant to be complete, nor should it be taken as such. It is provided as a guide to point out some areas of safe practice and perhaps serve as a stimulant to enable the reader to reflect upon, and conjure up, other pertinent safety practices.

To supplement the efforts of the worker, as well as maintain the control achieved by the safety-design features, requires a program that can evaluate the effectiveness of the control mechanisms. This latter need is supplied by the health-physics program. Even though highly sophisticated instruments are used, and techniques are refined to the utmost detail, events that result in the loss of control will still occur. To mitigate the impact of

these events by limiting the extent of the loss of control quickly is a function also supplied by the health-physics program. The features of such a program in regard to plutonium work will be covered in this section. In addition, some aspects of fire and nuclear safety will also be discussed.

B. Contamination Control

The toxic nature of plutonium requires that this substance be kept out of the body. Since plutonium in air tends to form a finely divided, loosely held oxide, there is a danger of this material being dispersed into air and remaining airborne.⁵ The presence of such oxide in air creates an inhalation hazard. To prevent this, the plutonium is confined. However, the enclosure then becomes highly contaminated with the loose matter, so that a negative pressure must exist in the enclosure to avoid contamination leaks. Then, the airflow in the area will pass through the enclosed system and out through the filters in the exhaust system. This arrangement prevents the spread of contamination. However, since no system is perfect, leaks may still occur.

Since one must have access to the plutonium in gloveboxes, rubber or neoprene gloves are provided. These gloves protect the worker from contamination, but are subject to breakdown. This is one reason why surgical gloves are also worn: They provide another means of protecting the individual from contamination. In the event of a slight breakdown of the rubber gloves, the individual may not know this merely from appearance. Some other means must then be used to detect the spread of contamination.

Besides the two examples given above, loss of control may also occur as the result of operator error. Then again, an incident may be due to some unforeseen reason or unknown cause, but loss of control will still be the result.

In hood work, surgical gloves are worn to protect the worker's hands from contamination. If at all possible, one should not handle objects within the hood with the gloves directly. Something else, such as tissues, should be used. Then, the tissue becomes contaminated first, and the gloves act as a second line of defense. Hands should not be pulled out of the hood unless gloves are removed, since this may cause a loss of control. Also, if the gloves pick up significant contamination, they should be changed and disposed of as waste. The use of highly contaminated gloves only disperses the material throughout the hood and increases the chance of hand contamination if the glove fails.

1. Philosophy of Contamination Control

The goal of any contamination-control program is to limit the spread of contamination to as small an area as possible.⁵ This aids in reducing the exposure potential of the incident, eases the task of controlling the extent of the event (spill), and enhances cleanup of the area. One common approach is shown in Fig. 8:

- a. Set up zones or areas to isolate the more hazardous processes. Allow work with radioactive materials only in the laboratory area.
- b. Control access. Set up a security guard (G) at the entrance, or require a special badge to actuate the door lock.
- c. Direct traffic patterns. Permit access to the laboratory area only through the doors at either end of the area (E) and past monitoring stations (M).
- d. Set up monitoring (M) and/or decontamination stations (D) at zone interfaces.
- e. Use protective clothing that is limited to the zone. Leave clothing worn in the laboratory area in the change room (CR) when leaving the laboratory area.
- f. Insist that anything leaving the hazardous zone be surveyed for radiation.

Fig. 8

Design for Contamination Control

The above scheme provides a means to detect and limit contamination that might otherwise be tracked or transferred to other areas. The entrance may also be marked with instruction for use of monitoring devices and special protective equipment while in the radiation zone. Such devices or equipment may be provided there or in the change room. In some cases, decontamination stations located at the zone boundary may be desirable. All these features may be set up, but depend upon the good will of the workers if they are to succeed. That is, unless the workers, and the visitors to the area, realize the necessity for checking everything that leaves the hazardous area, the contamination-control concept will fail. The scheme to contain any contamination within the laboratory area in Fig. 8 works well only if the total cooperation of all personnel involved is obtained. When cooperation breaks down and exceptions to the control procedures begin to increase in frequency,

then the probability of losing control is greatly increased. On the other hand, with strict adherence to the control procedures, the extent of any contamination can be limited to the hazardous zone alone. In addition, if frequent surveys of the laboratory area are performed, contamination incidents are picked up more quickly. This also aids in reducing the size of the area involved in a contamination incident.

2. Contamination-control Practices

In addition to the practices discussed in Sec. IV.A above, the worker should observe other control measures. These measures seek to reduce the hazard potential at the local level as well as prevent intake of contamination. The use of shoe covers, laboratory coats, and surgical gloves that are not worn out of the area will help contain the spread of contamination. Survey of surgical gloves for radiation each time the hands are removed from the glovebox permits early detection of loss of control. The forbidding of smoking, eating, or drinking in the radiation area will reduce the chance of radioactive matter being inhaled or ingested. Hands should always be washed following work with radiation. When leaving a radiation area, one should monitor his hands, shoes, and any equipment he is taking to further prevent the chance of spreading any contamination. Wounds or punctures should always be monitored for plutonium and promptly treated by a doctor.

Waste-disposal practices set up for different forms of contaminated waste should be observed. Dry waste is often put in metal containers, sealed in plastic bags, and removed through bag ports.⁵ This waste may then be placed in large, covered, metal drums, which can be sealed when filled, and removed for burial. Liquid waste is handled by piping to waste tanks or containers, by use of small bottles or other containers, and by treatment (evaporation or absorption in vermiculite) that changes the liquid waste to solid waste.⁵ Gaseous waste is often planned for in the design features of the exhaust system.^{5,7} Pyrophoric wastes are often burned within an enclosure before being disposed of.

Waste-handling rules should specify the correct method of disposal for items that do not fit standard containers. Also, the correct use of waste containers as well as proper waste segregation should be explained.⁷

Plutonium storage practice involves nuclear-safety aspects that require special attention.⁷⁹ For this reason, storage must be strictly controlled. Large amounts of plutonium are often stored in areas or vaults of special design, and under the control of a responsible individual. He must keep records of the amount on hand and be responsible for proper storage and movement of plutonium. For safety purposes, access to these areas should be restricted.

For smaller amounts, the user should submit a material-handling plan that should include storage plans. He is responsible for the movement and

storage of this material in his area. This plan should be reviewed for approval in regard to nuclear safety. For amounts below an established minimum, the threat of a criticality is no longer a problem. However, depending on the form of the plutonium, special storage precautions may need to be taken. Such things as proper containers or storage in an inert atmosphere will warrant some thought.

3. Control Problems

The previous discussion was concerned with the general design and administrative aspects of contamination control. However, the problems in degree of control are specific to the material which is involved. For example, see Table XIV. This table groups together a number of materials that are presently in use. Included in the table are the ICRP values of the MPC in air and the body burden values, in both microcuries and micrograms, for each of the nuclides.

TABLE XIV. Occupational MPC and Body Burden for Selected Nuclides

Nuclide	MPC, $\mu\text{Ci}/\text{cm}^3$	Body Burden	
		μCi	μg
^{239}Pu	2×10^{-12}	0.04	0.65
^{238}Pu	2×10^{-12}	0.04	0.0023
^{252}Cf	6×10^{-12}	0.01	0.000018
^{210}Po	2×10^{-12}	0.03	0.0000067
Nat. U	7×10^{-11}	0.005	7400

If one compares the MPC values, the relative control problems would be about equal for all but natural uranium. This is also true if one compares body-burden values in microcuries. However, on the basis of MPC values, natural uranium appears to be 35 times less hazardous than ^{239}Pu ;

and on the basis of body burden in microcuries, eight times more hazardous. To assess the degree of control needed, one must look at the mass of material as well as the activity. That is, in a loss-of-control incident, the mass of any of the above materials that leaks out will be about the same, but the relative hazard this represents will differ greatly.

For example, assume 0.1% of a sample leaks out in an incident. If the sample were 650 μg of ^{239}Pu , the release would be 0.65 μg or one body burden. The same conditions involving natural uranium would result in $0.65 \mu\text{g}/7400 \mu\text{g} = 8.8 \times 10^{-5}$ body burden. If the incident involved ^{252}Cf , then $0.65/(1.8 \times 10^{-5}) = 3.6 \times 10^4$ body burdens would be released! Clearly, the degree of control needed in each case is vastly different. So, with respect to design, one must be aware of the added confinement needed on the basis of mass considerations.

4. Health-physics Control Measures

Health-physics control measures may be grouped into two classes: (1) Provide fixed monitors for detection of hand, shoe, clothing, or air contamination; and (2) provide portable instruments and monitoring service on a routine basis to maintain contamination control.

a. Fixed Monitors. A fixed monitor may be used to trigger an alarm when the measured radiation field exceeds a preset level. In this sense, the device indicates that the loss of control is significant. A fixed monitor may also be used to check the contamination status of an item. In this application, the device may indicate either no contamination or some level of contamination. Both types of devices may be used for certain operations.

(1) Personnel Monitor. A personnel monitor is an ac-operated device used to survey hands, shoes, and portable objects.¹⁰³ It features a chamber in the form of a flat plate-type probe of large area through which the counting gas passes. Alpha particles enter the chamber through a thin Mylar window and ionize the chamber gas. The chamber output, which is proportional to the alpha activity, is displayed on a count-rate meter and indicated by audible clicking from an attached speaker. The same device may be used for beta monitoring if the operating voltage is changed.

This type of monitor can be placed both at the enclosure and at exits to the work areas. When a hand is withdrawn from the enclosed system, it can be surveyed immediately without the risk of tracking any contamination. Upon leaving the work area, a worker is also able to survey his hands, clothing, and shoes. By making frequent surveys, he will quickly detect the presence of contamination. This action will limit the spread of any contamination.

Another type of personnel monitor, the hand-and-foot counter, is also used at the entrance to radiation areas. Such units may be designed to monitor both hands and shoes during the same counting period without requiring the use of a probe.¹⁰⁴ At the end of a fixed counting time, an indication of contamination or no contamination is given. When contamination is indicated, the level can then be measured with portable health-physics instruments. These monitors are useful when a large number of people require hand-and-shoe surveys.

(2) Continuous Air Monitors. Continuous air monitors are used to maintain a watch on the level of air activity in the work area. These instruments are usually of homemade design and consist of some air-collecting device and a suitable detector for the radiation.¹⁰⁵⁻¹⁰⁷ For gases, a flow-through chamber may be used to collect and detect the presence of the radioactive gas. For particulates or dust, filters are often used to collect the material, and the type of radiation emitted will dictate a choice of detector. The device is often provided with an alarm, since the presence of an airborne toxic substance may represent a severe inhalation hazard.

The presence of natural airborne radioactivity [see Sec. IV.B.4.b(4)] below] affects the ability of an air monitor to detect airborne plutonium. These radioactive products give rise to short-lived alpha and beta activities that tend to mask out any activity due to plutonium in an air sample. In the past, the sensitivity of air monitors has suffered because of this feature.

The continuous air monitor, CAM-5, designed at ANL,¹⁰⁵ counts both the beta and alpha activities built up on a fixed filter. The counting rates for each type of activity are then displayed on their own charts. Although the separate counting rates may vary up or down, the ratio of these counting rates will remain fairly steady if no other radioactive substance is present. This ratio is balanced out by electronic means to give a nearly null reading, which is displayed on a third or center chart. Unless some other activity is collected on the filter, the ratio remains fairly constant. Hence, the presence of an alpha emitter deflects a needle to one side of the central chart, and the presence of a beta-gamma emitter deflects the needle to the opposite side. This design greatly increases the sensitivity of the air monitor in regard to plutonium. However, the detector is not shielded so that external beta-gamma fields can adversely affect its operation.

(3) Stack Monitors. This type of fixed monitor is used to estimate levels of radioactive substances which are released to the environs. In case of an accident, it also serves to denote if control by the filter system has been breached and what the extent of the release may be. These types of monitors are often homemade devices that are designed and assembled for specific jobs.

b. Monitoring Services. The monitoring services provided by an active health-physics program include routine instrument and smear surveys for surface contamination, personnel and item contamination surveys, and standby monitoring. Air sampling of the general work area, as well as special samples during certain operations, are also carried out.

(1) Surface-contamination Surveys. Routine surface surveys should include both instrument and smear surveys. The frequency of these surveys will depend upon the form of the plutonium and the type of operation carried out. In most cases, because of the degree of confinement used for plutonium, a positive survey result is cause for concern. That is, even a rather low level of contamination should warrant both cleanup and further investigation. For this reason, surface-contamination guides or levels are not readily found for plutonium.¹⁰⁸ At any rate, the toxic nature of plutonium is such that any loss of control should mandate followup surveys. In some cases, certain contamination levels may be tolerated on the basis that proven control techniques are able to maintain contamination at or below these levels.

A number of portable or survey instruments are used to perform routine surface surveys.^{25,103,109,110} Normally, alpha survey instruments are used for surface surveys when plutonium is involved. If the sample contains fission products, though, a beta-gamma survey instrument may be used. Gas-flow proportional counters¹⁰³ are used for all types of alpha monitoring. When humidity may vary greatly, these are preferred over air counters. The counter is a small, flat, platelike probe through which the gas flows. The alpha radiation enters the counter through a thin Mylar window and ionizes the

chamber gas. The counting rate displayed on the instrument meter is proportional to the activity of the alpha source. Because of the short range of alpha particles, the probe must be used very near to the surface being surveyed ($<1/4$ in.). Earphones should be used with the counter to increase detection sensitivity. The lower limit of detection is about 250 dpm in the area under the probe.¹⁰³ This type of counter may be modified so that low-energy beta particles can be detected.¹¹⁰ This latter capability is useful in monitoring for some of the transplutonic radioisotopes. Alpha scintillation counters using ZnS crystals are also used for alpha surveying.^{25,109,110} Gas-flow and scintillation counters both respond to neutron fields, so that extra care is needed when surveying for alpha surface contamination.

The smear survey consists of wiping a filter medium or other suitable material across a surface that is suspected of being contaminated.⁵ The smear can then be surveyed for activity with a portable instrument or counted for activity in a laboratory counter. This technique is easy to apply and allows quick spot-checking of areas for loose contamination. In many cases, one is concerned more about the presence of the contamination rather than its level. The qualitative results obtained in smear surveys are most useful in these instances. The method is also used to check the integrity of clad or sealed sources to ensure that no leakage has occurred.

(2) Personnel and Item Surveys. In addition to the fixed monitors used for surveys in the work area, many other surveys of both personnel and items are made by survey instruments. These surveys are required when transfers into and out of enclosures are made. Surveys are also required for such things as skin punctures or wounds, glove changes on enclosures, decontamination of personnel or items, and repair work on potentially contaminated objects.

(3) Standby Monitoring. In some cases, the nature of the work may be hazardous enough to require the presence of a health physicist during the entire process. This is called standby monitoring. This type of monitoring is also good practice each time a new technique is being used. The advantage here is that the worker can devote full concentration to the job and the health physicist will take care of the necessary monitoring. Also, the worker has the benefit of health-physics advice at each step of the job.

(4) Air Sampling. Air sampling is carried out to assess the condition of the work-area environment. It also allows a check on the effectiveness of the control design and/or work practices in regard to contamination control. Since internal dosimetry is difficult to perform, common practice has been to limit the concentration of radionuclides in air. Although the MPC values in Table XII are used as standards that should not be exceeded, the goal is to keep airborne levels as low as possible. The MPC value (or CG value of ERDAM 0524) is used as an index of control, and airborne concentrations well below this level imply satisfactory control. To confirm that this is the case,

routine air samples are collected. When air samples reveal levels above the standards, this is viewed as a potential breakdown of the control system. Levels greatly above the MPC values for short periods do not constitute a severe hazard of themselves, but such levels over extended periods would be of concern. In the former case, it is not so much the level but the apparent loss of control that should be the major concern. Once control is lost, levels may go even higher as more material is released.

1. Natural Airborne Radioactivity. Airborne radioactivity has always been present as a part of man's environment. It arises largely because of the presence of uranium and thorium in soil throughout the world.^{25,55} The long-lived isotopes ^{238}U and ^{232}Th have long decay chains, which produce many daughter products. In each chain, a noble-gas daughter product is born: radon (^{222}Rn) in the ^{238}U chain and thoron (^{220}Rn) in the other. These inert gases migrate from the soil to the air, forming, along with their short-lived daughters, the major portion of natural airborne radioactivity.

The daughter products of these gases are particulate matter and attach to the dust in air. These are then captured by air sampling, and their activity often overwhelms that of the isotope of interest when the sample is counted shortly after sampling. This interferes with the proper interpretation of the air sample results.

2. First-count Factor. One scheme in use at Argonne to deal with the problem of natural airborne radioactivity is the "first-count factor"²⁵ or alpha-beta-gamma factor.¹⁰³ This factor is obtained from the ratio of beta-gamma to alpha activities on the air sample, determined as quickly as possible after sampling ends. This factor is remarkably constant when only the radon-thoron daughter products are present.²⁵ If the ratio varies dramatically upward, the release of a beta emitter is signaled, and conversely, a major downward shift implies an alpha release. The actual value of the first-count factor is a function of many parameters and may vary from system to system. However, an air-sampling history of an area will quickly establish this number for a given system for the case of natural air activity. Once the factor is known, one can use the initial counting results of the air sample to estimate if an activity release has occurred.

3. Air-sampling Practices. The most common means of sampling for particulates is to collect them on filters.²⁵ The sampling technique is to draw air at a known flow rate through a filter for a known length of time. Figure 9 depicts a typical system.

Fig. 9
Schematic Diagram of
Typical Filter Sample

Once the sample is collected, the filter can be removed and counted for both alpha and beta activity on separate counters. After the activity is measured, the concentration may be computed, since the air volume is also known.

Samplers may be located in the general work area to routinely monitor the overall operation. In other cases, samplers may be positioned close to a source of potential air contamination, such as hoods or gloveboxes.⁶⁴ Personal air samplers¹¹¹ have also been used to obtain air samples in the breathing zone of the individual. Thought must be directed to the type of sample needed in regard to sampler placement. Large differences have been obtained in the results of some of these methods for sampling a given atmosphere.^{100,111,112} The frequency of air sampling in an area will depend upon the form of the plutonium, the nature of the work, and the presence or absence of fixed air monitors.

C. Internal Exposure Control

1. Personnel Monitoring Measures

There are many problems in assessing the uptake, distribution, and retention of plutonium and the subsequent internal exposure that results.^{63,113-120} In Secs. III.B.1 and III.B.2, many of these factors were briefly discussed. To estimate the internal-exposure rate, a measure of the organ burden is needed. Given the organ burden and excretion data, the total dose equivalent may be estimated. The control methods discussed attempt to exclude the entry of radioactive matter. Nevertheless, such entry may occur, so that methods are needed to estimate the burden and subsequent radiation-dose equivalent.

A common method used to appraise internal exposures is a bioassay program. Routine urine and/or fecal samples are collected at some frequency, reduced chemically, and counted for the plutonium content. Soluble plutonium retained in the body is excreted mainly through the urine, whereas insoluble plutonium is found mostly in the feces. Data obtained from a number of samples can be used to estimate organ burdens.^{63,121} In the event of a suspected intake, it may be necessary to collect all excreta for the first few days following an accident.¹¹³

In recent years, whole-body counters of special design have been used to detect plutonium in the body.^{114,115,117} These devices are able to detect the 17-keV X rays from ²³⁹Pu and the 60-keV X rays from ²⁴¹Am. Since these X rays are easily absorbed in dense substances, the method is useful for lung-burden determinations, but not for bone burdens. To correctly assess the lung burden by this method requires that the ²³⁹Pu/²⁴¹Am ratio be known.^{115,122} This method, used in conjunction with bioassay samples, has improved the estimate of intake in the case of inhalation events.¹¹⁸

Wound monitors have been developed that can be used to estimate the amount of plutonium in wounds.¹²³ These instruments also count the low-energy X rays from the plutonium. These devices are useful for cases in which plutonium may be embedded in the tissue. Then, alpha radiation will be totally absorbed and not detectable, whereas the X rays can still be seen. These monitors aid the physician treating the wound since the plutonium should be removed and may have to be excised. Normal skin contamination by plutonium can be detected by portable alpha survey meters.

On occasion, other samples such as nose swabs, sputum, or blood may be needed in order to help in assessing the intake.⁷

2. Therapeutic Measures

A number of substances have been found useful for increasing the urinary elimination of plutonium from the body.^{63,70,103,116,124} Of these, diethylene-triamine-penta-acetic acid (DTPA) has proven the most effective agent.⁷⁰ However, treatment with this substance is most effective only in the early stages following intake. It is presumed that once plutonium is bound in bone, DTPA will have little effect.¹²⁰ In the early stages, when the level of soluble plutonium in the blood is high, DTPA is able to increase the excretion rate. When plutonium is removed, the subsequent total dose may be greatly reduced. This feature is most important since soluble plutonium is so tightly bound in bone, ensuring long, continuous irradiation.

In recent years, pulmonary lavage (lung irrigation) has been tried as a means of removing inhaled matter from lungs. One application of this method to humans has been reported.¹²⁵

3. Protective Equipment

Protective equipment should be supplied to the worker to increase the degree of protection afforded by other safety measures. Its purpose is to protect him from contamination and to aid in the control of contamination.¹²⁶ The use of such equipment guards against pickup of external contamination on the worker's person or clothing and intake of contamination into the body. Also, the equipment is removed by the individual in the given area so that the contamination can be confined to that area.

Types of protective clothing used are laboratory coats, coveralls, and pants and shirts.⁷ Coveralls or pants and shirt are preferred, since these call for a change of personal clothing. This precludes loss of personal clothing in a contamination incident or spill. Gloves and safety shoes may also be provided to round out the basic scheme. In some cases, one may provide laboratory coats and shoe covers at the zone entry for use by observers or those not normally working in the area. Any handling operation with plutonium should be done with protective gloves on. Other more elaborate types of clothing may also be used.¹²⁶

Other necessary protective devices include respiratory equipment. These may be needed for special operations, such as repair work or modifications, as well as for an emergency. The use of these devices on a continuing basis in place of proper source control of the plutonium work is undesirable. One class of respirators, the full facepiece type, processes the air the worker breathes in from his surroundings.¹²⁶ These are suitable for particulate matter, and some may provide protection up to 100 times the MPC in air.¹²⁷ In the other class of respirators, which includes self-contained breathing units, the worker does not breathe the air from his surroundings. This type is suitable for both gases and particulate matter and should be used for particulate concentrations greater than 100 times the MPC in air.¹²⁶ Here again, the function performed and the form of the plutonium used will affect the type and required use of the above protective equipment.

D. External Exposure Control

In Secs. III.A.5.a and III.A.5.b, the external dose rates from a plutonium sample were discussed. At that time, expressions were presented that showed that the external field is a sensitive function of the isotopic composition. For plutonium that contains less than 5% on a weight basis of isotopes other than ²³⁹Pu, the external field is not a great problem.⁶¹ Above this weight percent, the external exposure hazards increase and eventually require special controls, such as reduced work time, shielding, or semiremote handling.^{29,61,99} Of course, recently irradiated uranium capsules may contain fission products in amounts comparable to the plutonium content. These would present very serious external exposure hazards that would require extensive shielding and totally remote handling, such as in hot cells. This latter consideration also applies to handling large amounts of transplutonium matter.⁹⁴

We saw, in the case of a plutonium sample, that the external field is comprised of low-energy X rays, low- and high-energy gamma rays, and fast neutrons. Beta radiation is absorbed either by the enclosure itself or by the enclosure gloves. X rays and gamma rays are reduced to some extent by the enclosure and by using leaded gloves. However, the photon dose rate increases with surface area of the plutonium. Thus, a thin layer of plutonium on the floor area of the enclosure may increase the photon dose rate. The presence of ²⁴¹Pu will mean a gamma-dose-rate (>40 keV) increase for years due to ²⁴¹Am buildup. Neutron dose rates depend upon the mass of the sample in the case of spontaneous fission. The dose rate from neutrons may also be greatly increased if low-Z-number substances are used in the process.²⁹

In the early handling of plutonium, extremity exposures were of little concern. With the advent of high-exposure plutonium, hand exposures as well as whole-body ones became important, and methods of reducing such exposures had to be provided.^{29,61} For transplutonium substances, dose rates are such that whole-body doses must be greatly reduced for all but small amounts of these substances.⁹⁴

1. Exposure-control Practices

Among the practices that may be used to limit the external exposure received by the worker are:^{61,94}

1. Limit the amount of material in the enclosed system.
2. Reduce the total work time allowed in handling plutonium.
3. Process the material as soon as possible after chemical separation.
4. Remove waste matter as quickly as possible so that surface deposits do not build up.
5. Use remote handling devices, such as tongs, as much as possible.
6. Rotate personnel for those jobs having the higher exposure rates.

2. Shielding

A number of substances have been used as shields to reduce the external radiation dose. Initially, the normal thickness of material on the enclosure provided enough shielding to reduce the low-energy X ray to low enough levels. As the plutonium isotopic composition shifted, the use of leaded gloves became common practice. These gloves were used to limit hand exposure, but their thickness, in lead equivalents, was restricted since at some point handling operations became too difficult. Common practice has been to limit thickness to 30-mil leaded gloves (0.1-mm lead equivalent). As both gamma and neutron dose rates became larger, it was necessary to go to more potent shielding to achieve needed dose reduction. In the case of gloveboxes, only so much shielding can be included or added and still allow ease of manual handling. Therefore, at some point, hot cells and remote handling devices must be used.

In the case of X and gamma rays, high-Z-number substances are most useful as shields. However, a shield material does not absorb equally for all energies of the incoming photons. The transmission expresses the fraction of the incoming photons of a certain energy that pass through a shield of given thickness. Table XV gives photon-transmission values for some common materials used in gloveboxes.

TABLE XV Transmission of X and Gamma Rays in Selected Materials⁷⁷

Material	Transmission			
	17 keV	43 keV	60 keV	100 keV
1/2-in. Lucite (Plexiglas)	0.28	0.74	0.81	0.82
30-mil lead gloves (0.1-mm lead equivalent)	0	0.38	0.67	0.84
1/4-in. aluminum	0	0.45	0.80	0.78
1/8-in. steel	0	0	0.046	0.43

As may be noted from the table, as photon energy increases, the shield thickness must also be increased to retain the same shield transmission factor. For transplutonic isotopes, which have more photons in the range above 100 keV than plutonium, the photon shielding problem becomes more troublesome.³⁹

In the case of neutron shields, the choice of a proper shield substance is more complex. For a fast-neutron spectrum, the shield must quickly remove the neutrons, yet be thick enough to absorb photons produced in radiative capture.¹²⁸ In very thick shields, the neutrons will be removed and still enough matter will be available to reduce the capture gamma ray hazard. In thin shields, one may reduce the neutron hazard but increase the gamma hazard.

For fast neutrons, low-Z-number substances, such as hydrogen, are most useful as shield materials.^{26,89,129} One may compare neutron-shielding substances on the basis of their hydrogen content. On this basis, polyethylene and water are among the better neutron shields, while concrete is one of the poorer ones.^{26,89} Reference 26 contains data on the shielding ability for some of these materials for fast neutrons. Transmission factors for thin neutron shields are difficult to obtain, since one must deal with a spectrum of neutron energies. In thin shields, the spectrum changes rapidly over short penetrations, so that the shield effect may depend upon the initial spectrum assumed. In thick shields, removal theory for neutrons may be applied.²⁶ Reference 130 contains shielding information useful for transplutonium material.

3. Health-physics Control Measures

Health-physics control measures cover monitoring services with survey instruments to define the radiation field and personnel monitoring programs to assess exposure. In addition to these features, fixed monitors that respond to external radiation fields above certain preset levels may also be used. The response may include visual and audible alarms to indicate accident situations requiring prompt action.

a. Monitoring Services. A number of survey instruments are used to estimate the external radiation field. For photons, beta-gamma survey meters (Geiger counters) or ion chamber devices are most often used.^{25,110} Neutron fields may be evaluated by devices that measure the neutron-flux density (such as the long counter²⁵) or approximate the dose equivalent rate (such as the neutron monitor¹¹⁰). Flux-density units may be related to the estimated dose equivalent rate by means of the relationships in Table XVI. These values have been adapted from Ref. 26.

For a fast-neutron spectrum, such as in fission or (α, n) processes, an average value for the flux density is used. The flux-density

value for a mean energy of 1 MeV is often used to estimate the dose equivalent rate. Any moderation of fast neutrons then provides a safety factor. This is particularly true when the field is measured after passing through hydrogen-rich shields.

TABLE XVI. Average Q Values, and Neutron-flux Densities That Deliver a Maximum Dose Equivalent of 100 mrem in 40 hr

Neutron Energy, MeV	Q (avg)	Neutron-flux Density, n cm ² -sec	Neutron Energy, MeV	Q (avg)	Neutron-flux Density, n cm ² -sec
Thermal	2	680	5×10^{-1}	11	27
1×10^{-2}	2	680	1	11	19
1×10^{-6}	2	560	2.5	9	20
1×10^{-5}	2	560	5	8	16
1×10^{-4}	2	560	7	7	17
1×10^{-3}	2	680	10	6.5	17
1×10^{-2}	2.5	700	14	7.5	12
1×10^{-1}	7.5	115	20	8	11

Estimates of the total dose rate from neutrons and gamma rays can be used to assess the hazard. If the dose rates greatly exceed the allowed standards, a severe hazard may exist. In these instances, actions that will greatly limit exposure during handling will have to be taken. For not so severe hazards, only minor changes may be called for.

b. Personnel Monitoring Devices. The most common type of device used to monitor the exposure of the worker is the film badge. These devices can be equipped with film that responds to beta, photons, and neutrons. After the badge is worn for a suitable period (2-4 weeks), the films are developed and the response can be related to the amount of exposure. A pocket dosimeter is often used with the film badge to allow visual checks of the exposure received.⁶¹ These devices can be read and recharged, and the reading recorded on a daily basis, if desired. Units can be used that respond to beta-gamma as well as to thermal and/or fast neutrons.

Film rings and wrist badges may also be worn for monitoring extremity exposures. In recent years, film has been replaced by thermoluminescent (TL) dosimeters in the finger rings.^{61,94} One reason has been that the film has been subject to light leaks, which produce erroneous response. This is not a problem with TL substances.

A problem in the use of film for neutron monitoring by track counting has been poor response in the intermediate-energy region.^{25,61,94} The NTA film in common use will not respond to neutrons below about 0.5 MeV. For moderated neutrons, this film may miss a large portion of the neutron dose. One method used to overcome this defect involves track-counting of fission-fragment tracks recorded in mica.¹³¹ The recoil tracks of fission

fragments, which arise from neutron capture in ^{235}U foils, appear as pit marks in the mica. Following an etching process, the tracks may be counted under microscope and related to the total number of neutrons to which the foils were exposed. Since ^{235}U fissions for all neutron energies, this method will cover the intermediate-energy region. This method has been in use at Argonne National Laboratory and has resulted in more realistic neutron exposures being found.¹³² Other methods^{61,94} have been used to correct for any loss of response in using NTA film.

E. Nuclear Safety

Nuclear safety may be defined as the prevention of uncontrolled critical-mass reactions.¹³³ For our purposes, this means preventing a criticality in any system meant to be subcritical. This task involves solving problems of a scientific, engineering, and administrative nature.^{79,134} Scientific problems involve the setting of values for the critical mass of the substance. Engineering problems involve process design to avoid conditions that favor a chain reaction. Administrative problems involve setting up practices, rules, and controls that attempt to preclude such occurrences due to errors in handling and processing.⁷⁹

Using the data from critical-mass studies, the process and equipment can be analyzed to arrive at safe mass limits throughout the plant. Then, detailed rules to achieve nuclear safety can be drawn up, and an administrative system of checks can be set up to ensure that these rules are followed.⁷⁹

1. Control Methods

The goal of nuclear safety is to minimize the chance that a criticality or excursion occurs. This goal is attained if a sufficiently large fraction of the neutrons released in fission are captured (nonfissile) or escape from the system (leakage). Then, fewer fissions occur in one generation than in the preceding one. A number of factors influence the neutron balance of the system so that several types of controls may be used.^{79,133,134} When these

factors are applied, the principle used is that nuclear safety should depend as little as possible upon actions or decisions by personnel.

Fig. 10. Example of Safe Geometry for Pipes or Cylinders

For solutions, safe geometry or "always safe shape" is the preferred method of control.¹³³⁻¹³⁵ In this method, vessel dimensions are such that enough neutron leakage occurs to prevent any excursion (see Fig. 10). The general approach is to limit one of the dimensions (such as the diameter of the cylinder in the figure) to ensure that enough neutrons will be permanently lost. Then, a chain

reaction cannot be achieved in the isolated system, regardless of the amount of fissile material. That is, the cylinder in the figure could be of infinite length and contain any concentration of fissile matter in the solution without going critical.

In many processes, the absence of substances that may moderate and/or reflect neutrons cannot be guaranteed. For example, the human body contains a great deal of hydrogen. For this reason, the values assume that a thick hydrogenous reflector surrounds the material. The thickness, diameter, or volume of vessels can be designed so that criticality cannot occur. This method may also be used for metals in the form of slabs, cylinders, and spheres (see Table XIII, recommended values).

Mass control is often the method used for metals if safe geometry cannot be used. The "always safe mass" is that amount of the fissile substance that cannot sustain a nuclear chain reaction, regardless of its shape or form and the nature of the surroundings. This mass, usually taken as slightly less than half the minimum critical value, is listed as the recommended value in Table XIII. This allows for one double-batching error, since two always safe masses will not cause a criticality. Mass control is often used for both glove-box and hot-cell work.^{11,134} The posting of the mass limit on the enclosure is a common practice that aids proper control practices.

Other methods of control, such as concentration limits, use of fixed and soluble poisons, and Pu/U ratio are discussed in the literature.¹³⁴⁻¹³⁶

In all cases, the limits used apply to isolated systems.

Fig. 11. Interaction Effects in Adjacent Containers

In the use and storage of fissile substances, the spacing must be taken into account to nullify any interaction between two systems.⁷⁹ In Fig. 11, container A is separated from container B by enough distance so that much of the n leakage from A is not intercepted by B, or vice versa. In this case, both containers A and B could be geometrically safe, as isolated systems. In the setup pictured, those systems would also be safe by virtue of the sufficient separation. However, the probability of interaction occurring is proportional to the solid angle, which increases with the decrease in the distance of separation. Thus,

if container B were at position C in the figure, more of the n leakage would be intercepted. This could produce a situation in which enough neutrons would not be permanently lost from the system of two containers. That is, enough neutron leakage from the entire system to avoid an excursion may not be obtained if the spacing of fissile matter is close enough. These neutrons may

cause fission in a nearby system, and neutrons from that system may do likewise. In this manner, two subcritical systems may be made critical if care is not taken.¹³³ Storage in arrays or cubicles, which have posted mass limits, is one means of obtaining the proper separation.

2. Administrative Controls

Administrative controls may require that a written nuclear-safety statement or manual be prepared for review by competent individuals if more than safe amounts are to be handled routinely.^{62,137} These statements should describe the processes, facilities, and equipment to be used as well as the makeup, masses, and configurations of the fissile substance.⁶² Credible accidents must be treated as well as safe procedures for double-batching or two simultaneous, unlikely events occurring.^{79,134}

For each approved statement, one individual should be responsible for seeing that only approved procedures are followed. Because of the importance of nuclear safety, the responsibility for nuclear safety must be clearly defined within the facility.⁷⁹ Detailed controls that are set up must be arrived at by each facility through analysis of unique functions.

3. Monitoring and Dosimetry Practices

Although the risk of an excursion is very low, it cannot be entirely eliminated, so that these events may accidentally occur.⁸⁶ For this reason, one should provide (1) a means of alerting workers to the threat of high-radiation fields and (2) a plan for their quick evacuation.⁸⁵ The exposures in such an event can be quite severe, so a delay of even a few seconds in leaving the area can be crucial.

The radiation alarm system should have a very rapid response and good reliability, give little or no false alarms, and emit a distinct, audible signal.^{81,85} This system should sound an immediate local alarm so that an area can be evacuated. It should also indicate the location and radiation level of the event at some central control station. The sensor, or radiation detector, may respond to either gamma or neutron fields. This is usually placed near the point where an excursion is a possibility. The alarm system, which may consist of a number of horns, may be distributed throughout the rest of the area. Personnel should be instructed so that they are aware of the alarm sound, the proper evacuation route, and the vital need for immediate evacuation. Drills and periodic training should be carried out to ensure that this information is not forgotten.

If a criticality occurs, a system is needed to obtain data on the absorbed dose to which a worker has been exposed. Such data will be of value for a number of reasons.¹³⁸ To obtain this information, we need a measure of both the neutron fluence and spectrum and the gamma-absorbed dose. Such

information can be obtained by the use of one of a number of nuclear-accident dosimeters.¹³⁸ Since placement of these fixed units may differ from the location of the exposed worker, a measure of the ^{24}Na activity induced in the person's body is also needed. With the ^{24}Na data, the dose at the dosimeter station can be related to the dose the worker receives.^{84,139}

The nuclear-accident dosimetry system in use at Argonne used a modified Hurst dosimeter package.^{140,141} The dosimeter is made up of a neutron section and a gamma section, which are physically tied to each other. The neutron section contains a bare gold foil, a cadmium-clad gold foil, a sulfur pellet, a cadmium-clad copper foil, and a ^{237}N , ^{235}U , and ^{238}U fission-foil system in a cadmium and ^{10}B shield. This unit measures neutron fluence in the thermal, intermediate, and fast regions to provide a rough spectrum estimate. This information allows the neutron dose to be estimated. The gamma dosimeter has chemical components and a TL dosimeter for measuring gamma dose.

To aid in dose determination, a "quick-sort" method is needed to roughly divide those exposed from the unexposed.¹⁴² A common method used is to provide the person with an indium foil. When exposed to thermal neutrons, the foil gives off beta-gamma radiation. This can easily be read with beta-gamma survey instruments if the check is made soon after the incident. An alternative method is to use a gamma-survey meter at the body midsection to measure induced radioactivity in the body.¹⁴²

In addition to the fixed dosimeters, the worker may be provided with neutron-measuring components that are part of his film-badge unit.¹⁰⁹ At ANL, these components include sulfur powder, an indium and two gold foils in a cadmium box, and a copper foil taped to the outside of this box. In addition to spectrum information, some information in regard to orientation in the neutron field can be obtained.

In principle, nuclear-accident dosimetry methods are simple and easy enough to apply. In practice, an accident may be a complex affair in which many unforeseen obstacles can appear.¹⁴³ These factors may delay or impede the dose determination in any given event. For example, a dosimeter may be destroyed or badly contaminated in the incident. Also, the radiation field may be so high that the unit cannot be readily retrieved. In these cases, other activated substances may be found that can be used to measure neutron activation.¹⁴²

F. Fire Safety

1. General Discussion

The most effective means of preventing plutonium fires is to use a dry, inert atmosphere, such as argon, helium, or nitrogen, in the enclosed system. Both moisture and small amounts of oxygen (see Sec. II.D) should be

avoided when pyrophoric metal is involved. For some operations, dry air has been used with success. Room air is used for aqueous solution lines and where clad material only is handled.⁵

Such design features as the use of noncombustible materials, fire breaks in an enclosure line, wire-mesh fire screens for exhaust filters, fire-detection systems, alarms and control for overpressures, fire-suppression systems, and other detection systems increase the degree of fire safety.^{5,7,11,12,88,144,145} Practices that control the sources of ignition and explosion treated in Sec. III.D will also help to prevent fires. Methods such as maintenance of airflow to prevent gas buildup, monitors for explosive mixtures, and control of inert-atmosphere purity aid in explosion control.^{11,12}

Fire safety in areas outside an enclosed system should be designed to protect the exterior of the enclosure from damage. The construction and fire-protection design of rooms and areas in which enclosures are found is important. These should be constructed of fire-resistant or noncombustible substances and provided with fire walls that isolate the area from other areas of the plant.⁵ The module, room, or building should remain standing to provide containment in the event of a serious fire in which material may be released from enclosures. Automatic as well as manual fire-fighting systems should be available.^{5,12} Automatic water-sprinkler systems can be used in areas in which there is no danger due to criticality or the presence of reactive metals (for example, sodium).⁵ Other types of area systems have also been developed.^{146,147} Portable or manual fire extinguishers should also be provided. These may include dry powder, CO₂ systems, fog nozzles, or foam devices, depending upon the nature of the fire hazard.^{5,12}

2. Fire Extinguishment

Since fires in enclosures such as hoods and gloveboxes can be expected, one should prepare for them. In addition to automatic systems, manual systems for fire extinguishing should be provided. Quick-connect couplings have been used on gloveboxes to ease the task of discharging portable fire extinguishers.⁵ Piercing lances or nozzles may be attached to extinguishers to pierce a glove in order to combat fires. Storage of manual extinguishing agents within the glovebox may be provided when an inert atmosphere is not used.

A number of reports have discussed the methods of extinguishing plutonium fires.^{5,7,43,44,148} In this respect, it is important to distinguish between fires that threaten to involve plutonium and those in which the plutonium is actually burning. If no plutonium is burning, a number of extinguishers are effective.^{5,7,44} Carbon dioxide (CO₂) devices are effective for flammable liquids and electrical fires, but will often pressurize the system.⁵ A dry-chemical type will safely extinguish all but metal and filter fires.⁴⁴ Water should be avoided for fires in which reactive or pyrophoric metals and/or fissile

substances are present. However, it should not be totally ruled out, even where these problems exist, since it may be the only means of controlling the fire.⁵³

For plutonium-metal fires, complete exclusion of oxygen and/or rapid heat removal to below the ignition temperature (500°C) are the only truly effective means to extinguish plutonium fires.⁴³ The work of Felt⁴³ showed that applying argon locally was very effective in extinguishing fires in large amounts of plutonium metal. However, one could not attain oxygen exclusion merely by flooding an air box with the inert gas. Only with a somewhat reduced oxygen content (<4%) could the argon stop the burning. This suggests either that an inert atmosphere be used or that some other method be available for extinguishing plutonium fires in an air atmosphere.

The use of fusible salts or metal powders does not exclude oxygen, but these substances are effective in removing heat. Of course, when the temperature drops below the ignition point, the metal no longer supports combustion. Based on the heat-removal aspect, solid agents that were most effective were magnesium oxide (MgO) sand and copper powder. Other agents tested were graphite, ternary eutectic salts, lead powder, iron powder, foam, plutonium dioxide, and Halon 1301 gas.⁴³

Sometimes plutonium metal fires treated by the above agents may not be extinguished easily. The main concern is to contain the plutonium fire and prevent it from igniting other substances. Often the greater hazard occurs when other combustible substances are ignited by the metal fire.^{43,44} When this happens, the fire can spread very rapidly throughout the system and great damage may occur before control can be regained.

V. EMERGENCY PLANS

Much has been said in regard to preventing certain incidents, yet they still seem to occur. A number of causes can be assigned or postulated in each case, but the cogent feature is that, despite all that can be done, accidents still occur. For this reason, one must accept the premise that certain accidents are going to occur. To properly deal with these situations at any level requires both a well-thought-out plan and trained individuals to execute the details.¹⁴⁹ As one might expect, the more complex the incident, the more detailed will be the plan and the more substantial will be the group involved.

The philosophy that should apply in an emergency situation will reflect these goals:⁷

1. Protect personnel from injury and hazards, and quickly identify exposed personnel.
2. Avoid actions that might create greater hazards.
3. Bring the emergency under control.
4. Prevent property damage.
- ✓ 5. Limit the extent of any contamination.

To deal with an event and form a plan of action will require an initial analysis of the accident potential. Thought should be given to the type and nature of events likely to occur. These may include criticality accidents, contamination release in fires or explosions, or high-radiation fields.^{7,149} At this stage, extra precautions should be taken to reduce the likelihood of the event, if possible. One should ponder the extent of the area and estimate the number of people involved. Key areas should be identified, and photographs of the suspected trouble spots may be prepared. Other building data and plans to aid emergency response groups unfamiliar with the area may also be included.

Once the events are defined, written procedures for immediate actions in these emergency situations can be prepared. There may be many facets to the plan, depending upon the nature of the event. However, at every level these plans should (1) be easy to understand, (2) specify actions to be taken, (3) detail responsibilities, (4) define communication lines, and (5) denote sites of emergency equipment. The plans should cover such aspects as evacuation plans, reporting emergencies, and the types and meaning of alarm systems. Assembly points and decontamination centers, and the proper routes to these areas, should be detailed. Drills and training of personnel should be carried out to test response and familiarize workers with their part in these plans. Review of these drills should be carried out to point out weak spots in the scheme so that these may be corrected.⁸¹

When immediate local actions are not able to cope with the event, a major emergency plan should be provided. This will require the response or assistance of many groups: medical people, health physicists, communications personnel, security forces, firemen, utility men, and others. The major plan should designate a director whose function is to coordinate the actions to ensure a team effort. He should be assisted by other designated personnel whose responsibility is to provide expertise in certain aspects of emergency control. Included in this group should be someone familiar with the area in which the emergency has taken place.

Among the functions that must be carried out in the event of a major incident are:

1. Evaluation of the situation in regard to degree of hazard.
2. Assistance in the evacuation or recovery of persons.
3. Surveying of persons for contamination and/or radiation.
4. Accounting of persons involved in the incident.
5. Formulation of procedures to bring the emergency under control.
6. Treatment and decontamination of any injured persons.
7. Provision of instruments, clothing, portable decontamination units, and other specialized equipment.
8. Collection of personnel data and preparation of public information.

Following any incident, a review of the event should be conducted with a view toward attaining greater safety in the future. In addition, the emergency measures and response of emergency groups should be evaluated. This may indicate methods of improving the handling of future emergencies.

Since facilities may be vastly different, the details of an emergency plan must be worked out on an individual basis. The intent here is simply to point out some of the aspects that must be treated in any actual plan. Reference 150 discusses several aspects in regard to emergency planning.

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