BNL47594
INFORMAL REPORT

Received
JUN 23 1992

LECTURE NOTES FOR CRITICALITY SAFETY

by

Ralph Fullwood

March 1992

DEPARTMENT OF NUCLEAR ENERGY, BROOKHAVEN NATIONAL LABORATORY
UPTON, NEW YORK 11973

Prepared for the U.S. Department of Energy
Under Contract No. DE-AC02-76CH00016

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

NOTICE

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, or any of their employees, makes any warranty, expressed or implied, or assumes any legal liability or responsibility for any third party's use, or the results of such use, of any information, apparatus, product or process disclosed in this report, or represents that its use by such third party would not infringe privately owned rights.

The views expressed in this report are not necessarily those of the U.S. Department of Energy.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

BNL--47594

DE92 016009

LECTURE NOTES FOR CRITICALITY SAFETY

Prepared by:

Ralph Fullwood

March 1992

Engineering Technology Division
Department of Nuclear Energy
Brookhaven National Laboratory/Associated Universities, Inc.
Upton, New York 11973

Prepared for:

U.S. Department of Energy
Washington, D.C. 20585

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

20
21
22

ABSTRACT

These lecture notes for criticality safety are prepared for the training of Department of Energy supervisory, project management, and administrative staff. Technical training and basic mathematics are assumed. The notes are designed for a two-day course, taught by two lecturers. Video tapes may be used at the options of the instructors. The notes provide all the materials that are necessary but outside reading will assist in the fullest understanding.

The course begins with a nuclear physics overview. The reader is led from the macroscopic world into the microscopic world of atoms and the elementary particles that constitute atoms. The particles, their masses and sizes and properties associated with radioactive decay and fission are introduced along with Einstein's mass-energy equivalence. Radioactive decay, nuclear reactions, radiation penetration, shielding and health-effects are discussed to understand protection in case of a criticality accident. Fission, the fission products, particles and energy released are presented to appreciate the dangers of criticality. Nuclear cross sections are introduced to understand the effectiveness of slow neutrons to produce fission.

Chain reactors are presented as an economy; effective use of the neutrons from fission leads to more fission resulting in a power reactor or a criticality excursion. The six-factor formula is presented for managing the neutron budget. This leads to concepts of material and geometric buckling which are used in simple calculations to assure safety from criticality. Experimental measurements and computer code calculations of criticality are discussed.

To emphasize the reality, historical criticality accidents are presented in a table with major ones discussed to provide lessons-learned. Finally, standards, NRC guides and regulations, and DOE orders relating to criticality protection are presented.

CONTENTS

	<u>Page</u>
ABSTRACT	iii
LIST OF FIGURES	vii
LIST OF TABLES	viii
ACKNOWLEDGEMENTS	ix
INTRODUCTION	x
 1. NUCLEAR PHYSICS	 1-1
1.1 Physics Review	1-1
1.2 Nuclear Particles and Natural Radioactivity	1-4
1.3 Nuclear Reactions	1-8
1.4 Radiation Protection	1-12
1.5 Dose, Dose Rate and Health Protection	1-16
1.6 Nuclear Resonances and Nuclear Fission	1-20
 2. NUCLEAR CHAIN REACTIONS AND CRITICALITY	 2-1
2.1 The Principle of the Chain Reaction	2-1
2.2 Neutron Economy	2-1
2.3 The Six-Factor Formula	2-2
2.4 Neutron Control	2-3
2.5 Lethargy and the Average Logarithmic Energy Decrement	2-4
2.6 Nuclear Reactor	2-7
2.7 Time Response of a Nuclear Chain Reactor	2-9
2.8 Rules for Avoiding Criticality	2-11
 3. DETERMINING CRITICALITY	 3-1
3.1 Buckling	3-1
3.2 Measurement of Criticality	3-5
3.3 Subcritical Limits	3-8
3.4 Calculation of Criticality without a Computer	3-11
3.5 Computer Calculation of Criticality	3-17
 4. CRITICALITY ACCIDENTS AND THEIR PREVENTION	 4-1
4.1 Some Characteristics of Criticality Accidents	4-1
4.2 Selected Criticality Accidents	4-2
4.3 Summary Tables	4-9

CONTENTS (Continued)

	<u>Page</u>
5. STANDARDS, GUIDES AND ORDERS	5-1
5.1 ANSI/ANS Standards	5-1
5.2 Nuclear Regulatory Commission Regulations and Guides	5-8
5.3 Department of Energy Orders	5-10

LIST OF FIGURES

	<u>Page</u>
1-1 A Cross Section Target	1-8
1-2 Isotropic Radiation	1-12
1-3 Fission Cross Section of U-235 from 0.01 to 5 eV	1-20
1-4 Five Stages in Liquid-Drop Breakup	1-21
1-5 Fission Product Mass Distribution	1-21
1-6 Fission Neutron Energies	1-22
 2-1 How a Reactor Works	 2-1
2-2 Six factors about the Critical Cycle	2-3
2-3 Neutron-Proton Collision	2-4
2-4 Thermal Energy Spectrum	2-4
2-5 Simplified Reactor	2-7
 3-1 Slab Flux Profile	 3-3
3-2 Critical Mass and Radius for a Bare and a Reflected Plutonium- Solution Sphere	 3-8
3-3 Multi-Parameter U-235 Spherical Sub-Criticality Limits	3-9
3-4 Multi-Parameter U-235 Slab Sub-Criticality Limits	3-10
3-5 Extrapolation Lengths for Cylinders Containing UO_2F_2	3-11
3-6 Extrapolation Lengths for Cylinders of Uranium Metal and δ -phase Plutonium	 3-12
3-7 Figure 3-3 Modified to Include Mass-Concentration Equation	3-13
3-8 Birdcage Shipping Container	3-14
 5-1 The Ring of ANSI/ANS Standard Protection	 5-2

LIST OF TABLES

		<u>Page</u>
1-1	Everyday Nuclear Particles	1-5
1-2	Q-factors for Various Radiations	1-17
1-3	Q-factors for Neutrons	1-17
1-4	Some Maximum Doses from NCRP-91	1-18
2-1	Moderating Properties of Several Materials	2-5
2-2	Delayed Neutrons from Uranium-235 Groups	2-10
3-1	Bucklings for Regular Geometries	3-3
3-2	Single Parameter Limits for Uniform Nitrate Aqueous Solutions of Fissile Nuclides	3-7
3-3	Single Parameter Limits for Metal Assemblies	3-7
4-1	Process Accidents	4-11
4-2	Bare and Reflected Metal System Accidents	4-12
4-3	Moderated Metal and Oxide System Accidents	4-13
4-4	Fissile Solutions and Miscellaneous System Accidents	4-14
5-1	Some 10CFR Titles Applicable to the NRC	5-8
5-2	Some Division 3 Regulatory Guides Relevant to Criticality	5-9
5-3	Some Parts of DOE Order 5480 Relevant to Criticality	5-10

ACKNOWLEDGMENTS

I gratefully acknowledge the assistance of Luther Lowry of Lawrence Livermore Laboratory who kindly provided video tapes and many booklets used in criticality training at his Laboratory.

Dr. John Carew, the BNL Criticality Safety Officer, very kindly and ungrudgingly provided his time and advise in the preparation of this course.

I also wish to thank: Jerry C. Cadwell for his many helpful suggestions in preparing this course; Dr. Avril Woodhead for the effective technical editing and help in radiation effects; and Susan Monteleone for the document preparation.

INTRODUCTION

An accident resulting in an uncontrolled nuclear chain reaction is called a "criticality accident." We will learn to recognize and calculate what materials, in what quantities, combinations, and shapes can result in a chain reaction¹. We will learn how to protect ourselves and others if such a reaction takes place. We will discuss the few serious and few fatal accidents that have occurred to show the reality of the criticality, and to gain lessons-learned to prevent criticality. Finally, we will review the ANSI/ANS standard, NRC Regulatory Guides and DOE Orders for preventing criticality accidents.

The following are the chapters to be covered:

1. Nuclear Physics,
2. Nuclear Chain Reactions and Criticality,
3. Determining Criticality
4. Criticality Accidents and their Prevention
5. Standards, Guides, and Orders

Chapter 1 - Nuclear Physics provides an understanding of the particles involved, radioactive decay, reactions, cross sections, and radiation protection by distance, shielding, and reduced exposure time. Measures of radiation and the effects of radiation on humans are presented, followed by discussions of nuclear resonances, fission, and the energy distribution of the fission neutrons.

Chapter 2 - Nuclear Chain Reactions and Criticality covers the principle of the chain reaction, the six-factor formula for calculating the neutron multiplication (k-factor), how to control criticality, the moderation and reflection of neutrons, how a reactor works, and ends with a discussion of the time response of a reactor.

Chapter 3 - Determining Criticality begins by showing that four of the factors in the six factor formula are equal to k_{∞} - the neutron multiplication factor for an infinite reactor. The other two factors are related to the Fermi age, the thermal diffusion length, and buckling. Buckling, related to the size of the reactor, can be used to determine if an assembly or process is critical. Next, there is a discussion of the experimental measurement of criticality and critical facilities. Single parameter and multiple-parameter limits that have been determined are presented, and used for the hand-calculation of criticality by four methods. The chapter ends with a discussion of the two types of computer codes for calculating criticality.

¹ A chain reaction is a succession of fissions occurring one after the other like a chain - the preceding event causing the following.

Chapter 4 - Criticality Accidents and their Prevention begins by discussing the explosive and lethal effects of criticality accidents. Sixteen accidents are discussed, one of which was a chemical explosion. One criticality accident, Chernobyl, killed more people than all of the 41 criticality accidents that are summarized in four tables.

Chapter 5 - Standards, Guides, and Orders end the course with the rules that have been developed, based on experience and theory, to prevent criticality accidents.

1. NUCLEAR PHYSICS

1.1 Physics Review

The following is a review of definitions and principles, presented in alphabetical order, that are commonly used in criticality safety.

Atomic Mass Unit - AMU

One AMU = 1/12 the mass of a carbon 12 neutral atom. This value is the standard for comparison of all of the other atomic masses. $1 \text{ AMU} = 931.494 \text{ MeV} = 1.66054\text{E-}27 \text{ kg}$.

Atomic Number - Z

The charge of the nucleus in multiples of the elemental charge; the number of protons in the nucleus.

Atomic Weight - A

The weight of an atom in multiples of an atomic mass unit (AMU)

Atoms

Atoms are constituents of molecules. They are composed of a central nucleus where practically all of the atom's mass is concentrated and planetary-like electrons circling the nucleus. Atoms exchange electrons with other atoms to produce the force binding atoms to form molecules.

Electron

The electron is the nuclear particle that flows in wires all about us to power homes and industry. Electrons are negatively charged, very light-weight particles, that circle the positively charged atomic nucleus to neutralize the overall atomic charge. If there are more or less electrons than are needed to balance the nuclear charge, the atom is said to be "ionized."

Electron Volt

Charged particles acquire energy when they pass between two electrodes of different potential according to the formula: $E = e \cdot V$, where E is the energy, e is the elemental charge (discussed below), and V is the accelerating voltage. Thus, energy is proportional to, and may be measured by, voltage instead of its fundamental and very large unit, Joules. This unit, called electron-volts is related to the Joule as:

$$1 \text{ Joule} = 1.602\text{E-}19 \text{ electron-volt (eV)} \quad (1-1)$$

Elemental Charge

Electrical charge comes in multiples of the elemental charge which is:

$$e = 1.602\text{E-}19 \text{ Coulombs (C)}. \quad (1-2)$$

Electrons are negatively charged; protons are positively charged, but the amount of charge carried by either particle is the same, regardless of sign.

Electric charge must be conserved

The sum of the electric charges before a reaction must be the same as the sum of the charges after a reaction.

Energy-Mass Equivalence

Einstein discovered that mass and energy are related through the famous equation:

$$E = m \cdot c^2, \quad (1-3)$$

where m is the mass, and c is the velocity of light. That is, if an amount of mass m vanishes, an amount of energy given by equation 1-3 must be released. This relationship is used to express nuclear mass in units of energy.

Isotope

Nuclei are called isotopes if they have the same number of protons but different numbers of neutrons. For example, ^{235}U and ^{238}U are isotopes. Both have 92 protons but the former has $235-92 = 143$ neutrons, while the latter has 146 neutrons. Chemically they are identical, because chemical properties are the result of the number of protons - however, nuclear properties may be quite different.

Neutrons

One of the constituents of a nucleus is the neutron, a particle having no charge and weighing 1836 times the electron weight.

Proton

The other nuclear constituent is the proton, having the same amount of charge as the electron but of opposite sign. It also weighs 1836 times the electron weight.

Review Questions:

- 1) What particles are nuclear constituents?
- 2) What do protons and electrons have in common?
- 3) What do protons and neutrons have in common?
- 4) What is an isotope?
- 5) What units are used to express the energy of molecules, atoms and particles?
- 6) How can the mass of a nucleus be expressed in units of energy?

1.2 Nuclear Particles and Natural Radioactivity

1.2.1 Summary of Particles

Scientists in the DOE complex working with very high energy accelerators produce exotic, rapidly decaying particles that are not relevant to criticality safety. This discussion is limited to what I call the "everyday" particles.

The 1903 Nobel prize was awarded to Henri Becquerel and the Curies for the discovery of radioactivity in certain uranium and radium bearing ores. In a magnetic field, the radiation was found to be composed of three types of particles which they identified by the equivalent of A, B and C except they used Greek letters:

A. Highly ionizing heavy particles having a positive charge called the "alpha" (α) particle, later identified as the nuclei of helium.

B. Lightly ionizing light weight particles with a negative charge called the "beta" (β) particle, later identified as an electron.

C. Lightly ionizing particles not affected by the magnetic field called the "gamma" (γ) particle, and later identified with electromagnetic, x-radiation.

Before this and subsequently, other particles of concern to us were discovered. Table 1-1 summarizes the everyday particles, classifying them by mass and charge; to simplify our discussion, they have been numbered.

1. Electron (e^-)

Electrons are the particles circling the atomic nucleus, having a mass of 9.108×10^{-31} kg, and a charge of -1.602×10^{-19} C.

2. Positron (e^+)

The positron is the mirror image of the electron, with a mass of 9.108×10^{-31} kg, and a charge of $+1.602 \times 10^{-19}$ C. The positron is the anti-particle of the electron according to Dirac's relativistic quantum mechanics. Every particle has its anti-particle with which it may annihilate e.g., $\beta^+ + \beta^- \rightarrow 2\gamma$. The energy of annihilation is contained in two identical gamma rays each having 0.511 MeV emitted in opposite directions to carry off and conserve the energy and momentum.

3. Proton (p)

The proton is a hydrogen atom without the electron i.e. the nucleus of the hydrogen atom. The proton has a mass of 1.672×10^{-27} kg, which is 1836 times heavier than an electron. The

charge is the same as the electron but of opposite sign.

4. Neutron (n)

Similar to the proton in that both constitute the atomic nucleus. Its mass is 1.6725×10^{-27} kg, and it has no charge.

Table 1-1 Everyday Nuclear Particles				
	Name	Mass (MeV)	Charge	Decay
1.	electron, β^- , beta minus	0.51099	neg.	stable
2.	positron, β^+ , beta plus	0.51099	pos.	anti-electron hence annihilates with electrons
3.	proton, p	938.27	pos.	stable
4.	neutron, n	939.56	none	888 s when free
5.	gamma, γ , photon	none	none	stable
6.	neutrino, ν_e	none	none	>300 s
7.	deuteron, d	1875.6	pos.	stable
8.	triton, t	2809.	neg.	12.3 y $H^3 \rightarrow He^3 + \beta^-$
9.	alpha, α	3727.4	2*pos.	stable

5. Gamma (γ)

The gamma or photon is an electromagnetic wave particle with no mass.

6. Neutrino (ν)

The "little neutron" has no charge and no mass but it is different from a γ -ray. It was originally postulated to exist to conserve energy, charge, and some other quantities in the beta decay of radioactive material. It is very hard to detect because it can penetrate great thicknesses of material without interacting. Its existence has been established in many experiments since its existence was postulated by Pauli in the early 1930s. There are several types of neutrinos: the one listed in Table 1-1 is the one that accompanies beta decay.

7. Deuteron (d)

The deuteron is the nucleus of deuterium which combines with oxygen to make heavy water. It is not an elementary particle because it is composed of a proton and a neutron. Its weight is not the sum of the neutron and proton weights because of the binding energy. It has a mass of $3.344\text{E-}27$ kg, and one unit of positive charge.

8. Triton - t

This nucleus of tritium is not an elementary particle because it is composed of two neutrons and one proton. Its mass is $5.0058\text{E-}27$ kg, with one elemental positive charge.

9. Alpha - α

This nucleus of tritium is not an elementary particle because it is composed of two neutrons and two protons. Its mass is $1.10367\text{E-}26$ kg, with two elemental positive charges.

1.2.2 Radioactive Decay

Radioactive isotopes are distinguished by the rate with which they decay and by the type of particles emitted. Some isotopes decay much faster than others. Even the same isotope may emit one type of particle faster than another.

Experimentally, it is observed that the rate, dN/dt , at which particles are being emitted is proportional to the number, N , of nuclei that can emit particles. Writing this statement as an equation:

$$dN/dt = -\lambda * N, \quad (1-4)$$

where λ is the proportionality decay constant, and the negative sign means that N is diminishing with time, t . Equation 1-4 is easily integrated to give:

$$N = N_0 * \exp(-\lambda * t), \quad (1-5)$$

where N_0 is the number of radionuclides at time $t=0$.

If we ask, how long does it take for half of the material to decay, we set the ratio $N/N_0 = 1/2$ and solve for time; the result is called the half-life.

$$t_{1/2} = \ln(1/2)/\lambda = 0.693/\lambda. \quad (1-6)$$

The rate of radioactive decay is expressed in units of halflife. and may be found in the Chart

of the Nuclides². To use such data in equation 1-5, it is necessary to determine the decay constant (λ) using equation 1-6.

Why Express the Decay Constant in Half-Lives?

Equation 1-5 is easily calculated with a hand calculator if the decay constant is known. However, calculating the decay using half lives can be done by simple multiplication and division. Suppose a radionuclide has a 2-year half-life and you want to know how much is left after 8 years. Then $8/2 = 4$ half-lives are of concern, so $\frac{1}{2} * \frac{1}{2} * \frac{1}{2} * \frac{1}{2} = (\frac{1}{2})^4 = 1/16$, and only 1/16-th of the material is left after 8 years.

Review Questions

1. What is the mass of an electron in kilograms? What is its weight in pounds?
What is the mass of a positron?
2. What do gammas and neutrinos have in common?
3. What is the energy of each gamma if an electron and a positron annihilate?
4. Tritium has a half-life of 12.26 years. If a nuclear bomb is filled with 4 grams of tritium, how much is left after 24.52 years if there are no leaks and no refilling?

² "Nuclides and Isotopes", General Electric Company, Nuclear Energy Operations 175 Curtner Ave. M/C 684, San Jose CA 95125.

1.3 Nuclear Reactions

Collision Mean Free Path Length

If a particle reacts with a nucleus, the particle may be deflected, it may lose energy by exciting the nucleus, or the nucleus may be transformed. The likelihood of a particle doing any of these things is expressed in the units of area and is called the cross-section. Suppose we have a beam, like a flashlight beam, of particles hitting a target, how many reactions will take place per second? The converse problem is, what fraction of the incident particles pass through the target unaffected? These are the subjects of this sub-section.

Figure 1-2 shows a slab of material that is the target for our beam of particles. The target has dimensions of L_1 by L_2 , giving an area A , the thickness is dx , which is so thin that each nucleus is exposed to the beam. The figure shows an incident particle and a target nucleus in grazing incidence i.e. the centers of the two particles must come within a distance d or else there is a miss. This is true for all angles about the collision circle that has a circular area:

$$\sigma = \pi \cdot d^2, \quad (1-7)$$

where $d = r + R$ with r being the radius of the projectile and R being the radius of the target particle or nucleus.

The units used for cross-sections are the barn defined to be:

$$1 \text{ barn} = 1\text{E-}24 \text{ cm}^2. \quad (1-8)$$

The name is said to have arisen from the expression "big as a barn." It is large in terms of nuclear sizes, but it is a convenient area for expressing nuclear cross-sections.

Figure 1-1 A Cross Section Target

presented by nuclei to the total area, hence:

$$p = \text{Area of nuclei and particles/Target area} = n \cdot \sigma \cdot dx, \quad (1-9)$$

where n is the number of nuclei per unit volume (number density).

The probability, p of a single particle colliding is just the ratio of the area

The probability that N particles penetrate to depth x of material, where dN collide in the thickness dx equals the ratio dN/N as N becomes large. Then equation 1-9 becomes:

$$dN/N = -n \cdot \sigma \cdot dx. \quad (1-10)$$

The ratio, dN/N , may be called the differential transmission, dT . Equation 1-10 is solved by integration to give the total attenuation to a depth x as:

$$T = \exp(-n \cdot \sigma \cdot x), \quad (1-11)$$

where $\exp()$ is the base of natural logarithms raised to the power of the contents of the parentheses.

Notice that if $x=0$, then $T=1$ to indicate complete transmission.

Conversely we may define attenuation as one-minus-transmission:

$$A = 1 - T. \quad (1-12)$$

Using equation 1-11:

$$A = 1 - \exp(-n \cdot \sigma \cdot x), \quad (1-13)$$

which is zero for zero thickness as you might suspect.

To use equations 1-12 or 1-13, the number of nuclei per unit volume, n is needed.

The number of atoms in a gram-atomic weight is Avogadro's number $A_0 = 6.02E23$. The atomic weight, A is the mass in grams per gram-atom so A_0/A is the number of atoms per gram. The density ρ is the mass grams per cm^3 . Combining this information, the nuclear density is:

$$n = \rho \cdot A_0 / A. \quad (1-14)$$

Microscopic Neutron Cross-Sections

The cross-section we have been discussing is called the microscopic cross-section, σ . It is the area presented by a nucleus to a neutron of a certain energy for producing a given type of reaction.

A primary collection of data on the microscopic cross section of neutron projectiles incident on isotopic targets is the "barn book" (BNL-325). To find the attenuation of neutrons by material of thickness x , multiply the cross-section in barns by n , given by equation 1-14. Since Avogadro's number is $6.02E23$ and a barn is $1E-24$, the result of the multiplication

is 0.602 to give the attenuation as:

$$A = 1 - \exp[-0.602 \cdot \rho \cdot \sigma(\text{barns}) \cdot x(\text{cm})/A]. \quad (1-15)$$

Be sure the cross-section selected is that for the correct type of reaction, neutron energy, and isotopic composition.

Macroscopic Cross-Section

The microscopic cross-section usually occurs in combination with n , hence is convenient to define the macroscopic cross-section as the product of n times σ or:

$$\Sigma = n \cdot \sigma = n \cdot \rho \cdot A_0/A. \quad (1-16)$$

This has the units of 1/length and is convenient because with it, equation 1-11 becomes:

$$T = \exp(-\Sigma \cdot x). \quad (1-17)$$

The values of Σ for elements, including uranium, are tabulated for various energy groups³.

Mean-Free Path Length

Attenuation is also expressed in terms of the "mean-free path length", symbolized by $\langle x \rangle$ - the average penetration distance which is:

$$\langle x \rangle = 1/(n \cdot \sigma) \quad (1-18)$$

substituting into equation 1-11 gives:

$$T = \exp(-x/\langle x \rangle). \quad (1-19)$$

Thus, the mean-free path length is the thickness of material that reduces the transmitted radiation by "one-eth" ($1/e$), where $e = 2.71828$, or 36.8%.

Half-Thickness

The thickness to attenuate the radiation by one-half is called the half-thickness. It is related to the mean free path length as:

$$x_{1/2} = 0.693 \cdot \langle x \rangle. \quad (1-20)$$

³ For example, see H. Etherington, editor, Nuclear Engineering Handbook, McGraw-Hill, NY, 1958.

The Advantage of Using the Half-Thickness

The use of the half-thickness, like using half-lives, simplifies calculations. Radiation that penetrates one half-thickness of target is $\frac{1}{2}$ of the original amount. After another half-thickness that $\frac{1}{2}$ is reduced by another $\frac{1}{2}$ and so on.

Numerical Example of a Calculation Involving Half-Thickness

Suppose water of thickness 2 cm will attenuate neutrons by 2. How much will 6 cm of water attenuate neutrons? The 6 cm thickness is 3 half-thicknesses so the attenuation is $\frac{1}{2} * \frac{1}{2} * \frac{1}{2} = \frac{1}{8}$.

Review Questions

1. What is a cross-section?
2. How big is a barn?
3. If a neutron cross-section is 10 barns, the material density is 1 gm/cm³, the atomic number is 10, and the thickness is 3 cm, what is the attenuation?
4. If the macroscopic cross-section is 0.5/cm and the thickness is 2 cm, what is the attenuation?
5. What is a half-thickness?
6. If a material is 10 cm thick and the half-thickness for radiation is 2 cm, what is the attenuation?

1.4 Radiation Protection

Radiation protection is the use of materials, distance, and limited exposure times to protect personnel, equipment, and the environment from radiation.

1.4.1 Geometric Shielding

Figure 1-2 Isotropic Radiation

Point Source

Dose is related to the amount of radiation absorbed by people or equipment. Figure 1-2 shows a point radioactive source radiating S particles per second equally in all directions: it is called an isotropic source and hence, is suited to spherical geometry. If all S particles pass through this sphere in one second, the surface flux density (SD), the number of particles incident upon an area, is:

$$SD = S \cdot t / 4 \cdot \pi \cdot r^2, \quad (1-21)$$

where t is the duration of the radiation. The total radiation incident on the area is:

$$D = SD \cdot \text{area} = \text{area} \cdot S \cdot t / 4 \cdot \pi \cdot r^2 \quad (1-22)$$

The difficulty comes from the fact that the areal surface in equation 1-21 is curved on a sphere of radius r , cast by the outline of the object being irradiated. If this area is small compared with the radius of the sphere, it may be approximated as the flat area, in which case, the solid angle is:

$$\Omega = \text{area} / r^2. \quad (1-23)$$

Substituting equation 1-23 into 1-22:

$$D = \Omega \cdot S \cdot t / (4 \cdot \pi). \quad (1-24)$$

This equation shows that the radiation incident is proportional to the solid angle, the source strength, and the time duration of exposure.

If the area nearly equals r^2 , it may be calculated by integrating the very small area shown in Figure 1-2:

$$\text{Area} = \int r^2 \sin \phi \cdot d\theta \cdot d\phi. \quad (1-25)$$

Substituting equation 1-25 into equation 1-23, we find the solid angle can, in general, be calculated as:

$$\Omega = \int \sin \phi \cdot d\theta \cdot d\phi / (4 \cdot \pi). \quad (1-26)$$

An Example of Calculating the Interception of Radiation

Suppose you are 1.8 m tall and 0.4 m wide, your area is $1.8 \cdot 0.4 = 0.072 \text{ m}^2$. If you are standing 10 m from a source emitting $1\text{E}10 \text{ n/sec}$, then the flux density is: $SD = 1\text{E}10 / (4 \cdot \pi \cdot 100) = 8\text{E}6 \text{ n/m}^2 \cdot \text{sec}$. Because your area is 0.072 m^2 , the number of neutrons hitting your body is $0.072 \cdot 8\text{E}6 = 5.7\text{E}5 \text{ n/sec}$.

Doubling your distance from a point source reduces the exposure by 75%.

Geometric Shielding - Line Source

The effect of a line of radiation sources can be easily calculated if the radiation from a point source is known by simply adding the effects of a number of point sources along a line. In doing this the point source solution is called a Green's function and the combined effects is an integral over the Green's function. Without giving the full mathematical details, the attenuation from a line source is:

$$SD(a) = SD(b) \cdot (b/a) \quad (1-27)$$

This equation indicates that the flux density from a line source is inversely related to the distance from the source, where a is the distance to the object being irradiated, and b is a reference distance, with $SD(b)$ being the flux density at b. Doubling the distance from a line source reduces the exposure by 50%.

Geometric Shielding - Plane Source

If the source of radiation is uniform over the plane, the surface density is:

$$SD(a) = SD(b) \cdot \ln(b/a). \quad (1-28)$$

Thus the flux density from a plane source is attenuated as the logarithm of the distance from the plane - doubling the distance only cuts the dose by 31%.

1.4.2 Material Shielding

The discussion of geometric shielding ignored the effects of materials between the object and the source of radiation. Equation 1-11 showed that:

$$\text{transmission} = 1 - \text{attenuation} = \exp(-x/\langle x \rangle) \quad (1-29)$$

where x is the thickness of the shielding and $\langle x \rangle$ is the mean-free path length of the shielding material for the type and energy of the radiation. Now, we put this into equation 1-22 to get:

$$SD = S \cdot \exp(-x/\langle x \rangle) / (4\pi r^2). \quad (1-30)$$

which shows that radiation is attenuated both by the distance from the source, and by the shielding material.

1.4.3 Buildup Factor

Equation (1-30) is based on the assumption that if a radiation particle impacts a nucleus, it disappears. The collision processes depend very much on the type of particles involved in the collision. Heavily ionizing particles such as, alpha particles or protons, are very easily stopped by a small amount of material because they leave a dense trail of ions. They are not generally removed by a single collision but slowed with energy going into the ionizing process. On the other hand, electrons scatter off other electrons, and in this process, lose energy and produce a gamma. Subsequently, the gamma may react with another electron to produce an electron and gamma. This process is called a gamma cascade which is complicated to calculate. Neutrons may suffer many elastic collisions with nuclei, in a similar cascade process, until their energy is too low for this gamma cascade to occur. In addition, neutrons may inelastically scatter from nuclei to produce gamma rays or the neutrons may be captured. If they are captured to produce fission, on the average 2.5 new high energy neutrons will be produced.

These complex processes are simply treated as an attenuation process with a build-up factor in equation 1-30 to correct for the effect of these secondary effects. This buildup factor, $B(E,r)$ corrects equation 1-30 for the cascades as:

$$SD = S \cdot B(E,r) \cdot \exp(-r/\langle r \rangle) / (4\pi r^2). \quad (1-31)$$

Large computers calculate theoretical models of the secondary processes to produce tables of build-up factors. These tables are prepared for neutrons and gammas of various energies in many geometries and material combinations. Tabulated buildup factors depend on the type of primary radiation, the energy, E , of the primary radiation, the charge, Z , atomic number, A , and thickness of the shielding material.

A Numerical Example of a Build-Up Factor

For example, a 1 MeV point isotropic source of gamma-radiation has a buildup factor of

2.1 when penetrating a mean-free thickness of water. If the build-up factor is ignored (set to one), equation 1-33 shows that $\exp(-1) = 0.36$, or 36% of the radiation passes through the shield. But when the buildup factor is included, $2.1 \times 0.36 = 76\%$ of the radiation penetrates the shield.

Summary of Shielding

The calculation of shielding and the attenuation it provides is a complex problem involving transport theory. The complexity includes a geometric effect, exponential attenuation, and a buildup factor. The attenuation may be approximated, but usually requires recourse to tables of buildup factors for the type of radiation, energy, shielding material, and penetration depth. Shielding geometries may be complex, such as labyrinth corridors or ducts, so there will be no straight-line paths. Shadow shielding may be used such that workers are in the shadow of the shield. Monte Carlo computer codes are required to calculate such complex problems accurately.

Review Questions

1. What is the solid angle of a 1 m^2 area, 10 meters from a point?
2. If an isotropic neutron source is emitting $1\text{E}10$ neutrons/s, and you are 1.8 m tall and about 0.6 m wide, how many neutrons are hitting you per second.
3. Why is a build-up factor needed for shielding calculations?
4. How does the radiation vary with distance from point, line and plane sources of radiation?

1.5 Dose, Dose Rate and Health Protection

In this section, we will discuss how radioactivity, dose, dose rate, and health effects are defined. Then we will discuss how dose can be reduced by reducing exposure time, distance from the source, and by shielding materials between you and the source of radiation.

Units of Radioactivity

The activity of a sample, source or contaminated material is the rate at which radioactive disintegrations are taking place. The earliest term for this is the curie, defined to be:

One Curie is the amount of radioactive material that undergoes $3.7E10$ disintegrations per second.

The more modern unit is the Becquerel defined to be:

One Becquerel is the amount of radioactive material that undergoes 1 disintegration per second.

thus

One Curie (Ci) equals $3.7E10$ Becquerels (Bq)

Dose and Dose Rate

The underlying item of interest is the effects of radiation on the human body. These effects are related to the term "dose", which is a measure of the amount of energy deposited in an organ from the entry or passage of radiation. The rate at which dose is deposited in the organ is called the "dose rate". The usual situation is that radiation is being emitted at some rate (say x neutrons per second), and is producing a dose rate on organs or the whole body. The major effect on health is not the dose rate but the total dose received:

$$\text{dose} = \text{dose rate} * \text{time of exposure.}$$

Thus, dose may be reduced by a short exposure time even though the dose rate may be high.

Units of Dose

Dose comes from the energy deposited in tissue due to the trail of ions left by a radiation particle as it loses energy in traveling through material. In a gas, the charge from ionization can be collected as a measure of the energy deposited. Thus, the Roentgen was defined in terms of charge deposited:

Radiation	Q
Gamma rays	1
X-rays	1
Beta rays and electrons	1
Neutrons	10
Protons	10
Alpha particles	20
Heavy ions	20

Table 1-2 Q-factors for Various Radiations

One Roentgen is that amount of radiation that will deposit 1 esu of charge in one cubic centimeter of air at standard temperature and pressure.

This definition was not very useful because the primary interest is in the deposition of radiation energy in human tissue thus, giving rise to:

One Rem (Roentgen equivalent man) defined to 0.01 Joules per kilogram of tissue.

Recently, this quantity was redefined to be the "gray" which is:

One gray (Gy) is that amount of radiation that will deposit 1 Joule of energy in 1 kg of mass of tissue.

Around the beginning of this century, cancer and illness was discovered to be associated with excessive use of xrays. Mouth cancer in watch painters was associated with the use of radium in the paint. It soon came to be realized that radiation can have negative health effects.

Health Effects

Neutron Energy (MeV)	Q	Neutron Energy (MeV)	Q
2.5×10^{-8} (thermal)	2	5	8
1×10^{-7}	2	7	7
1×10^{-6}	2	10	6.5
1×10^{-5}	2	14	7.5
1×10^{-4}	2	20	8
1×10^{-3}	2	40	7
1×10^{-2}	2.5	60	5.5
1×10^{-1}	7.5	1×10^2	4
5×10^{-1}	11	2×10^2	3.5
1	11	3×10^2	3.5
2.5	9	4×10^2	3.5

Table 1-3 Q-factors for Neutrons

The concepts introduced as the rad (gray) do not adequately describe the impact of energy on tissue because absorption of a given amount of energy in a given mass does not describe the dose effect. Tissue damage increases with the linear energy transfer (LET) i.e. the density of the ionization along the track. The dose is estimated by correcting the gray by the "relative biological effectiveness" (RBE) or "Q" factor of various radiations relative to 200 keV x-rays.

Table 1-2 gives the Q-factors for several types of radiation; note that Q increases with LET. Neutrons and protons have similar LET because neutrons collide with protons.

Table 1-3 shows Q-factors for neutrons of various energies.

Table 1-4 Some Maximum Doses from NCRP-91		
Type of Exposure	Dose (mSv)	Dose (rem)
A. Annual occupational exposure		
1. Equivalent stochastic effects	50	5
2. Dose equivalents for tissues and organ (non-stochastic)		
a. Lens of eye	150	15
b. All others (e.g. bone marrow, breast, lung, gonads, skin etc.)	500	50
3. Guidance: cumulative exposure	10*age	1*age
B. Public (annual)		
1. Effective dose: continuous, or frequent	1	0.1
2. Effective dose: infrequent	5	0.5
3. Remedial action recommended when:		
a. Effective dose equivalent	>5	>0.5
b. Exposure to radon and its daughters	>0.007 Jhm ⁻³	>2 WLM
4. Dose equivalent limits for lens of eye, skin and extremities	50	5
C. Education and training (annual total)		
1. Effective dose	1	0.1
2. Dose limit for lens of eye, skin, and extremities	50	5
D. Embryo-fetus exposures		
1. Total dose equivalent limit	5	0.5
2. Dose equivalent limit in a month	0.5	0.005
E. Negligible individual risk level (annual) per source or practice (excluding background)	0.01	0.001

Health Effects: Dose Equivalent - the Sievert

Using the quality factor in conjunction with the energy deposited (the dose), we can estimate the health effects (H). This is expressed in the equation:

$$H = D * Q \quad (1-36)$$

where the dose equivalent is expressed in Sieverts and the dose is in grays. (100 Sieverts = 1 rem). It is important to note that the determination of dose equivalent requires knowledge of the absorbed dose, the composition of the radiation and its energy distribution.

Table 1-4 summarizes some guideline limits for radiological protection⁴.

⁴ From the National Council on Radiation Protection and Measurements, "Recommendations on limits for Exposure to Ionizing Radiation," NCRP report No. 91, 7910 Woodmont Ave., Bethesda MD, June 1987.

Review Questions

1. What is the difference between a Curie and a Becquerel?
2. What is the difference between Sievert and a rem?
3. Suppose thermal neutrons deposit a Joule in you body, and you weigh 70 kg. What is your dose in rem, and in Sieverts? Is this a significant dose?
4. If the dose in problem 3 was received in one hour, what was the dose rate?

1.6 Nuclear Resonances and Nuclear Fission

1.6.1 Nuclear Resonances

Fission is the splitting of a nucleus into two (rarely three) fragments of the original nucleus with the release of energy and particles. Fission occurs spontaneously in some isotopes heavier than uranium, but it usually occurs when a neutron is captured in a fissionable nucleus. The most common nuclei capable of fission are, U^{235} , U^{233} , and Pu^{239} (notice these are even in Z but odd in A).

Figure 1-3 Fission Cross Section of U-235 from 0.01 to 5 eV

The cross-section for a neutron being captured in a nucleus depends on the neutron's energy and the target nucleus. Nuclei are very complicated and are subject to resonances, shown by very large changes in the cross-section corresponding with small changes of neutron energy.

Figure 1-3 (left), from the barn book, shows the fission cross-section of U^{235} for incident neutrons from 0.01 to 1 eV; Figure 1-3 (right) shows the fission cross-section for incident neutrons from 1 to 5 eV. Figure 1-3 (left) clearly shows a general trend of neutron cross-sections changing in proportion to the reciprocal neutron velocity ($1/v$, where v is the neutron velocity). This, and the fact that neutrons accumulate in the thermal region where they enter into thermal equilibrium with materials, are the reasons that most reactors use thermal neutrons to produce fission.

The right part of Figure 1-3 shows the resonance structure which causes variations in the cross-section by nearly a factor of 100 with small energy changes. These resonances greatly complicate detailed calculations of criticality.

1.6.2 Nuclear Fission

A Liquid-Drop Model of Fission

The particular of reaction in which we are interested is the fission reaction. If a neutron is captured in a nucleus, the compound nucleus acts like a liquid drop in which the liquid

Figure 1-4 Five Stages in Liquid-Drop Breakup

begins to slosh and break-up into two or more large fragments - called fission fragments. Five stages from the initial capture of a neutron in a nucleus to the deformations becoming so severe

that the drop breaks up are shown in Figure 1-4.

Nuclear Fission

Figure 1-5 Fission Product Mass Distribution

The compound nucleus does not break into halves, but breaks into light fragment having mass about 95 AMU and heavy fragments having mass about 140 AMU. Figure 1-5 shows the distribution of the masses of the fission products.

Neutrons and other radiation are emitted during fission. The average number of neutrons emitted (called $\bar{\nu}$) is 2.55, 2.47, and 2.91 for thermal neutron fission of U^{233} , U^{235} , and Pu^{239} , respectively. The kinetic energy budget for particles from fission is: 165 MeV for fission fragments; 5 MeV for β s from fission fragments; 7 MeV for prompt γ s; 6 MeV for γ s from fission fragments; 10 MeV for neutrinos; and 5 MeV for neutrons, for a total of about 200 MeV.

Neutrons emitted from fission are distributed around 0.75 MeV, as shown in Figure 1-6. From Figure 1-3, it is apparent that the cross-section is lower at higher energies than at lower energies; therefore, fission occurs more readily at lower neutron energies.

Figure 1-6 Fission Neutron Energies

The distribution of the neutron energies coming from fission is shown in Figure 1-6. It should be noted that the peak of the distribution (most probable) occurs at about 0.75 MeV (this corresponds to a speed of 27 million mph).

Review Questions

1. What is the total amount of energy released in fission?
2. How much of the energy from fission is useful for the production of heat?
3. What is the fission cross section of U-235 at 25 meV (milli-eV - this energy is the average energy of room-temperature thermal neutrons).
4. Indicate the correct answer: Neutron cross-sections: a) are not affected by neutron energy, b) increase with increasing neutron energy, c) decrease with neutron energy in proportion to the reciprocal of the velocity.
5. What relatively plentiful isotopes are capable of low energy neutron fission.
6. Are the Zs of the isotopes of problem 5, even or odd (parity)? What is the parity of the As of problem 2?

Summary of Chapter 1

The nucleus of an atom is an extremely small central core containing 99.95% of the mass of the atom. The nucleus contains protons and neutrons. The chemical character that identifies it as an element is specified by the number of protons in the nucleus. The number of neutrons may vary to form different isotopic species of an element that are chemically the same but have different nuclear properties. One isotope may spontaneously decay into another or may de-excite by the emission of alpha, beta, gamma, neutron, neutrino particles, or it may undergo fission.

The rate of emission of particles is proportional to the amount of material decaying, hence, the decay is exponential with time. Radiation is attenuated by distance from the source of radiation; for a point source, it decreases inversely with the square of the distance. Radiation is attenuated by the presence of material between the object and the source; this attenuation is exponential with the thickness of material except for a correction factor called the buildup factor. The radiation is proportional to the source's strength (rate of particle emission).

Nuclear cross-sections express the area presented by nuclei as a target for being hit by elementary particles. The area is expressed in a unit called a "barn", defined to be 10^{-24} cm². Cross-sections expressed in these units are tabulated for neutrons impacting various isotopes. Penetrating ability also is expressed in half-thickness for ease of calculation. Neutron cross-sections vary greatly according to the isotopic target and with the neutron energy. There is a general "one-over-velocity" dependence, but at neutron energies above 1 eV, there are "resonances" in which the cross-section varies as much as 100 with small energy changes.

If neutrons are captured in certain odd-Z, odd-N nuclei specifically, U^{233} , U^{235} , and Pu^{239} , the nucleus undergoes fission with the release of a large amount of energy and nuclear particles.

This chapter has laid the ground work for the study of the combinations and shapes of materials necessary to produce a chain-reaction - called a criticality event.

2. NUCLEAR CHAIN REACTIONS AND CRITICALITY

2.1 The Principle of the Chain Reaction

Figure 2-1 illustrates the feedback process that occurs in a sustained chain reaction. At the top of the Figure, neutrons are born in the fissioning of a nucleus at an energy of about 0.75 MeV. At this energy, the fission cross-section is quite small but it increases generally as $1/v$. A criticality requires fissionable material, neutrons, and surrounding material to slow down the neutrons and to retain neutrons to continue causing fissions.

2.2 Neutron Economy

Whether an economy is growing, shrinking, or staying constant depends on the balance between expenditure and collection. The same is true of a reactor¹. A fission produces about 2.5 neutrons, on average. If only one of these neutrons is used to produce another fission, the reaction is self-sustaining and will continue until the fissile material becomes depleted.

The neutron multiplication about a fission cycle is designated by the parameter k , defined to be:

$$k = \frac{\text{Number of neutrons in one generation}}{\text{Number of neutrons in the previous generation}}$$

or:

$$k = \frac{\text{Neutron production rate}}{\text{Neutron loss rate}}$$

When: $k = 1$, a reactor is critical. It is neither gaining nor losing neutrons.

$k < 1$, it is losing neutrons

$k > 1$, it is supercritical.

Figure 2-1 How a Reactor Works

¹ In this and subsequent chapters, the word "reactor" means any configuration of materials capable of a chain reaction - not necessarily a device intended to become critical and generate power.

If $k = 1$, the reactor will continue at constant power; if $k > 1$ the power increases with each neutron generation. The bigger it is the faster the increase. If $k < 1$, the power decreases.

2.3 The Six-Factor Formula

The neutron multiplication is described by the six-factor formula which identifies the terms controlling the neutron economy:

$$k = \eta * \epsilon * p * f * p_{fnt} * p_{mt} \quad (2-1)$$

where,

- η (eta) - the number of neutrons produced in a fission to the number absorbed,
- ϵ - fast fission factor: ratio of the total number of neutrons produced by all fissions to the number produced by slow neutron fission ($\epsilon = 1.03$ for natural uranium),
- p - resonance escape probability: the probability that neutrons will escape capture and reach thermal energies where they may cause fission,
- f - thermal utilization: the ratio of thermal neutrons absorbed in the fuel to the total number of thermal neutrons absorbed,
- p_{fnt} - fast neutron nonleakage probability, and
- p_{mt} - thermal neutron nonleakage probability.

The first term, η , of equation 2-1 is a fact of nature concerning the fissionable materials selected. Similarly, the fast fission factor, ϵ is a function of geometry and the material. The remaining two factors may be varied by introducing neutron absorbing material. If the absorber absorbs neutrons at energies above thermal energies, then p , the resonance escape probability will increase. If it primarily absorbs neutrons at thermal energies, the effect will reduce p_{mt} , the thermal utilization; if absorption occurs at high energy, p_{fnt} will decrease. The use of moderators to slow and contain neutrons may affect either p_{fnt} or p_{mt} .

The six-factor formula is illustrated in Figure 2-2. At the bottom, 405 neutrons cause fission. As $\eta = 2.47$, 1000 neutrons are produced, some of which produce fast fissions ($\epsilon = 1.04$). In the slowing down process, 140 neutrons are lost from fast leakage, and 300 neutrons are lost from resonant capture. One hundred neutrons are lost by thermal leakage, 50 by non-fuel material absorption, 45 are lost by capture processes in the fuel that do not produce fission, and finally, 405 produce fission to start the cycle over.

A critical condition can be prevented or stopped by breaking this cycle.

We will now discuss in more detail the elements of the critical cycle.

2.4 Neutron Control

Neutron absorbers² may be liquids or solid materials.

Solid absorbers used primarily to stop a reactor are called scram rods; absorbers used to control a reactor are called "control rods". In both cases, the absorber may not be rod shaped³. Examples of materials used as the neutron absorber in control rods are: hafnium, europium, cadmium, and boron.

Solid absorbers may be built into a new core as "burnable poison" to help hold down the excess reactivity. As the core is used, the poison is designed to burn out to compensate for the reduction in fissile material.

Liquid reactor control is achieved by dissolving a neutron-absorbing material e.g. boric acid, in the cooling water. This is a slow control mechanism used in conjunction with control rods. When a core is new, and designed to last a long time, it has extra fuel. Liquid absorber is used to "hold down" the reactivity to a convenient level for automatic control by the control rods.

Gas absorbers are not in current use but there have been discussions at the Savannah River Plant and Oak Ridge National Laboratory on the use of the gas He^3 as an emergency shut-down mechanism.

If you have ever played pool, you will know that if the cue ball hits another ball dead-on, the cue ball may come to a dead stop and the other ball will go on. If the angle is a little different, both balls will continue moving but more slowly, and if the cue ball hits at a glancing angle, it is hardly affected. This is what happens if elastic balls of equal mass collide, and also happens if a neutron collides with a proton, such as in water.

Figure 2-2 Six-factors about the Critical Cycle

² A neutron absorber is an isotopic material with a large cross-section for absorbing neutrons without resulting in fission.

³ Fermi used cadmium sheets that dropped like a guillotine into the reactor if the "SCRAM" rope were cut. In current reactors, scram rods are moved in and out by the "Control Rod Drive Mechanism" (CRDM).

As a result of multiple collisions of the type shown in Figure 2-3, the neutrons lose energy and enter into thermal equilibrium like a "neutron gas". In many respects, the neutrons behave like hydrogen gas inside of the reactor materials. Thermal neutrons i.e. neutrons in thermal equilibrium with a reactor have a speed corresponding to the temperature of the reactor. The energy of these neutrons has a Maxwell-Boltzmann distribution as shown in Figure 2-4. The average energy of neutrons in this inverted "bell curve with a tail" is:

Figure 2-3 Neutron-Proton Collision

$$E(\text{eV}) = 8.61 \times 10^{-5} \cdot T(^{\circ}\text{K}). \quad (2-2)$$

At room temperature, 20°C (68°F), the neutron energy is 0.025 eV and its speed is 2200 m/s (5,000 mph) so the left part of Figure 1-3 encompasses the thermal distribution (except for a low energy tail).

The fact that the average energy of thermal neutrons is proportional to the temperature and the fact that neutron cross sections increase as neutron energy decreases explains why some solutions and reactors go critical when they cool-down.

Figure 2-4 Thermal Energy Spectrum

2.5 Lethargy and the Average Logarithmic Energy Decrement

If you average the energy loss of a neutron over the various possible scattering angles, a quantity called the average logarithmic energy decrement, ξ , may be calculated:

$$\xi = \langle \ln(E_1/E_2) \rangle \approx 2/(A+0.67). \quad (2-3)$$

The average number of collisions, n , needed for a neutron to go from 2 MeV to 0.025 eV are:

$$n = 18.2/\xi \quad (2-4)$$

Table 2-1 summarizes the properties of various moderating (slowing-down) materials in which the values of ξ and the number of collisions to thermalize are calculated on the basis of the lighter nuclei in the materials. Hydrogen, which is the primary agent for thermalizing neutrons in water, is very effective and can slow neutrons down, on the average, in only 18 collisions.

The value of ξ does not tell the whole story. Moderation also depends on the cross section for a collision. The product $\xi * \Sigma_s$ is called the "slowing down power" where Σ_s is the "macroscopic" cross section, which is:

$$\Sigma_s = \sigma * \rho * A_0/A \quad (2-5)$$

as derived in equation 1-16. The symbol, Σ_s , uses the s subscript to indicate that it is the macroscopic scattering cross-section i.e. neutrons elastically collide with nuclei but are not absorbed.

Table 2-1 Moderating Properties of Several Materials				
Material	ξ	Collisions to Thermal	Slowing Down Power (1/cm)	Moderating Ratio
water	1.0	18	1.53	72
heavy water	0.725	25	0.37	1.2E4
helium	0.425	43	1.6E-5	83
beryllium	0.209	86	0.176	159
carbon	0.158	114	0.064	170

In addition to these elastic properties, it is also necessary to consider neutron absorption in the moderator. This effect is accounted for by the moderating ratio defined as:

$$\text{Moderating ratio} = \xi * \Sigma_s / \Sigma_a \quad (2-5)$$

where Σ_a is the absorption cross section i.e. the cross section for a neutron to enter a nucleus with subsequent emission of something other than a neutron. In the process a neutron is lost. For the reason that heavy water is a poor absorber, it is an excellent moderator. Heavy water is the only moderator capable of producing a chain reaction in natural uranium, i.e. uranium as it is mined without enrichment. The reactors that work on this principle are the Canadian CANDU reactors.

Review Questions

1. What moderator can cause in a chain reaction in natural, unenriched uranium?
2. On the average, how many collisions must a fission-energy neutron make with hydrogen nuclei to reach thermal energy?
3. State the six-factor formula and explain its terms.
4. How much does the average thermal energy change when a reactor cools from 250°C to 28°C. (Remember to use Kelvin temperature.)

2.6 Nuclear Reactor

Figure 2-5 is a highly simplified diagram of a reactor such as a Savannah River Site production reactor. Neutrons produce fissions in the core which consists of fissionable

Figure 2-5 Simplified Reactor

uranium metal encased in aluminum tubes. Heavy water (D_2O) flows in the space between tubes to serve two purposes: remove the heat from the fissioning, and to moderate the high-energy neutrons from fission. Not all of the neutrons will be contained in the core region. To reduce neutron loss, the core region is surrounded by an additional D_2O moderator called the reflector. Its purpose is to reflect back, into the core, as many neutrons as possible - hence to keep the p^*f factors in the six-factor formula as close to 1 as possible.

The reactor is controlled by introducing neutron absorbing rods to reduce k by reducing the p^*f factors. Which factor is primarily affected will depend on the energy region in which

absorption takes place. The control rods are moved up and down by the control rod drive mechanisms. Some types of reactors dissolve a neutron absorber (such as boric acid) in the primary coolant/moderator.

The primary envelope consists of the pressure vessel (also called the reactor vessel), the pumps, and the piping that connects with the heat exchanger which is used to transfer the heat from the primary loop into a secondary cooling loop so that primary water which is slightly contaminated with radioactivity is not exposed to the environment.

A reactor is designed to go critical and operate with a power producing chain reaction. It is standard practice to enclose the chain-reacting core in an enclosure called the containment as shown in Figure 2-5 (although the Savannah River reactors were designed before this was the practice). The containment is the last line of the defense-in-depth policy of multiple barriers protecting the public.

Review Questions

1. What is the purpose of the reflector in a reactor? The fuel? The control rods?
2. The k factor of a reactor is slightly temperature dependent. Which way should it change for safety?
3. The coolant of a reactor serves two purposes. What are they?

2.7 Time Response of a Nuclear Chain Reaction

Prompt Period

The rate of disappearance of neutrons, N , from a reactor is proportional to the number of neutrons in it:

$$dN/dt = -N/T \quad (2-6)$$

where T is called the reactor period, which is the average time from fission to disappearance of the neutron either by escape, capture, or capture-fission; T will depend on the average neutron velocity and the size of the reactor. A fast reactor is characterized by the whole process from fission to fission capture taking place at high energy - in the range between a few MeV and a few keV. A fast reactor will have a T of, say 10 μ sec, an atomic bomb has a T of about 0.01 μ sec. A thermal reactor, such as a production reactor, has a T of about 0.001 sec. The number of neutrons, N , is related to the power, p , being produced by the reactor. With this substitution, equation 2-6 is easily integrated to give:

$$p(t) = p_0 \exp(t/T). \quad (2-7)$$

This equation shows that the power will increase by 2.71828 in a time $t=T$. For example, a thermal reactor will increase its power by 10,000 times in 1 second ($t=10 \cdot T$).

From this, it would seem that a reactor is difficult to control. Luckily, this is not the whole story, because some neutrons are delayed.

Delayed Period

But nature is not so perverse as to leave us in this situation. Instead, about 0.75% of the neutrons are delayed because they are associated with beta emission. As we saw in Figure 1-5, when a nucleus undergoes fission, it breaks into two parts centered about $A = 90$ and $A = 140$. For example, consider the fission reaction:

The first equation shows the fissioning of uranium-235 into lanthanum 147 and bromine-87 with two prompt neutrons (as required to balance the nucleons). Bromine-87 emits an electron with a decay constant of 55.6 sec. changing to krypton-87, which decays to krypton-86 by emitting a neutron. This is just one of the reactions that produce delayed neutrons. Table 2-2 summarizes the delayed emission as 5 groups.

It may seem that because the delayed neutrons are only 0.75% of the neutrons emitted that they are not significant. However if $k=99.25\%$ on the prompt neutrons, the delayed neutrons make the difference between being critical and subcritical. Thus controlling the reactor on the delayed neutrons raises the time constant to tens of seconds instead of milliseconds.

Prompt and Delayed Critical

An important distinction is made between $k \geq 1$ without including the delayed neutrons or including the delayed neutrons.

The former is called prompt critical. It is a run-away condition that usually is unstoppable. The reaction must go to completion, which usually means that so much heat and pressure is generated that the reactor disassembles itself. For example, it blows the liquid out of solution criticality or distorts metal parts to increase the neutron leakage sufficiently to stop. This whole process happens very fast. A solution, assembly, or reactor that is delayed critical means that the delayed neutrons which constitute 0.79% of all of the neutrons, cause $k \geq 1$. A small amount of absorber will shutdown the reaction and shut it down with a time constant corresponding with the delayed neutron times. All controlled nuclear reactors operate in the delayed critical region.

Table 2-2 Delayed Neutrons from Uranium-235 Groups			
Group	Half-life (sec)	Fraction	Energy (MeV)
1	0.62	0.00084	0.42
2	2.19	0.0024	0.62
3	6.5	0.0021	0.43
4	31.7	0.0017	0.56
5	80.2	0.00026	0.25

Review questions

1. What causes the neutrons to be delayed in being emitted?
2. Why are there five delayed neutron groups?
3. Thirty seconds after fission, what fraction of the delayed neutrons have been emitted?

2.8 Rules for Avoiding Criticality Accidents

The six-factor formula (equation 2-1) gives us guidance for avoiding criticality. Anything that breaks the critical cycle will prevent criticality. One method is to make it easy for the neutrons to escape by controlling the:

- Geometry.

For an assembly, or solution to go critical, it must be likely that a neutron born from fission will cause another fission. Neutrons from fission are prevented from escaping being surrounded by either more fissionable material or by material that reflects the neutrons back to fissionable materials for another try at fission. The geometry that does this best is the sphere⁴. Spherical geometries will go critical with less fissionable material than any other shape. Conversely, this is why fissionable material is stored in rod-like cylinders - to make it easier for the neutron to escape without causing fission.

An assembly whose shape is such that it cannot possibly go critical is called "geometrically safe."

Another method for controlling fission is:

- Neutron absorption.

Absorbing neutrons in a non-fission reaction is essentially the same as letting them escape. Criticality may be avoided by the use of such materials as boron (especially the isotope boron-10), cadmium or other materials used for scram or control rods.

Process streams may contain dissolved neutron absorbers (called "poisons") or absorbers may be build into the walls of the process containers - called rashig rings.

The speed on the neutrons, hence the fission cross section depends on:

- Temperature

Raising the temperature of an assembly, or process raises the average thermalized neutron energy reducing the fission neutron cross section, thereby increasing the possibility that neutrons will escape or be captured in a non-fission reaction.

Neutrons may be slowed by collisions with light-weight nuclei which increase the fission cross section. The slowing-down process is called:

⁴ A sphere has the least surface area for a given volume of any figure.

- Moderation

Light nuclei, including the human body are effective moderators. The presence of such material causes neutrons to be reflected back into the assembly and slows the neutrons. Both of these processes increases the reactivity and may make a subcritical assembly go critical.

The process of returning neutrons for another try at fission is called:

- Reflection

Materials that are good moderators are often good reflectors even though the principles of moderation and reflection are different. The purpose of a moderator is to slow neutrons; the purpose of a reflector is to cause neutrons to change direction.

Review Questions

1. Name 5 things affecting criticality.
2. What is the meaning of "geometrically safe"?
3. What is a good shape for geometric safety?

Summary of Chapter 2

Whether or not a reactor, assembly or solution goes or exceeds criticality depends on the k-factor.

- If $k=1$, the reactor is just critical i.e. as many neutrons are being generated as are being lost.

- If $k = k_{\text{prompt neutrons}} \geq 1$, the reaction is prompt critical and must stop itself;

- If $k = k_{\text{prompt neutrons}} + k_{\text{delayed neutrons}} \geq 1$ but $k_{\text{prompt neutrons}} < 1$, the reaction is delayed critical and is controllable by neutron absorption or leakage if done promptly;

- If $k_{\text{prompt neutrons}} + k_{\text{delayed neutrons}} < 1$, the reaction is sub-critical.

The 6-factor formula, used to calculate the k-factor, expresses the neutron budget about the chain reaction cycle. The six factors account for the number of neutrons produced by fission, additional neutrons from fast fission, the probability that neutrons are lost in slowing down from high energy, through the resonance region to reach and enter into thermal equilibrium. The six-factor formula accounts for neutron loss in the thermal region, non-fission capture, and for the number of neutrons born in fission.

This neutron slowing down process is natural and increases the likelihood of fission as slow neutrons are more likely to cause fission than fast neutrons. Light nuclei (small Z numbers) are used to slow down (moderate) the neutron energy to enhance the fission process.

The chain reaction takes place very rapidly, but some neutrons are released by beta-decay processes, having half-lives of the order of seconds. These delayed neutrons give the time needed to control a reactor.

Criticality accidents have occurred. Criticality may be prevented by interrupting the chain reaction cycle i.e., modifying terms in the six-factor formula.

3. DETERMINING CRITICALITY

Methods for calculating criticality are the subject of this chapter¹ which is intended to provide an overview to stimulate further.

3.1 Buckling

An Infinite Reactor

Chapter 2 showed there are reaction and escape processes taking place in a reactor. It is convenient to make this distinction in the six-factor formula by imagining a reactor so big that neutrons cannot escape, in which case: $p_{fml} = 1$ and $p_{mtl} = 1$. The six-factor formula then becomes:

$$k_{\infty} = \eta * \epsilon * p * f. \quad (3-1)$$

The six-factor formula may be written as:

$$k = k_{\infty} * p_{fml} * p_{mtl} \quad (3-2)$$

where, for convenience,

η (eta) - ratio of neutrons produced in a fission to the number absorbed,

ϵ - fast fission factor: ratio of the total number of neutrons produced by all fissions to the number produced by slow neutron fission.

p - resonance escape probability: the probability that neutrons will escape capture and reach thermal energies where they may cause fission,

f - thermal utilization: the ratio of thermal neutrons absorbed in the fuel to the total number of thermal neutrons absorbed,

p_{fml} - fast-neutron nonleakage probability, and

p_{mtl} - thermal-neutron nonleakage probability.

Age Diffusion Theory

Fermi solved the problem of neutrons slowing down from fission energies to thermal energies in terms of the age parameter τ , which is one-sixth of the mean square distance from the source to where the neutron reaches thermal energy, and the thermal diffusion length, L , which is one-sixth of the root mean-square distance that a neutron travels from where it became thermal to where it is captured. From this theory, the condition for criticality is when:

¹ The principal reference is R.A. Knief, Nuclear Criticality Safety, American Nuclear Society, La Grange Park, IL, 1991.

$$k_{\infty} \exp(-B_m^2 \tau) / (1 + L^2 B_m^2) = 1, \quad (3-3)$$

B_m is the material buckling that can be related to the six-factor formula by identifying the fast-neutron nonleakage probability to be:

$$p_{fnt} = \exp(-B_m^2 \tau), \quad (3-4)$$

and the thermal neutron nonleakage probability to be:

$$p_{nt} = 1 / (1 + L^2 B_m^2) \quad (3-5)$$

In the limit of large buckling, $\exp(-B_m^2 \tau) = 1 - B_m^2 \tau = 1 / (1 + B_m^2 \tau)$ and equation 3-3 becomes

$$k_{\infty} / (1 + B_m^2 (\tau + L^2)) = k_{\infty} / (1 + B_m^2 M^2) = 1, \quad (3-6)$$

where $M = \sqrt{(\tau + L^2)}$ is called the migration length. This is called the migration area method.

Geometric Buckling

After the neutrons enter into thermal equilibrium with the material, they act like a gas leaking from an enclosure - very similar to the problem of the temperature distribution in a hot block of material in which heat is flowing from the boundary to the surroundings. For neutron diffusion, the equation is:

$$\nabla^2 \phi + B_g^2 \phi = 0, \quad (3-7)$$

where ϕ (phi) is the neutron flux defined as the number of neutrons per unit volume times their velocity, B_g is the geometric buckling², and ∇ (del) is a partial differential operator used to mathematically describe diffusion from an enclosure.

For a slab, one dimensional reactor, equation 3-7 is solved as:

$$\phi = \phi_o \cos(x B_g), \quad (3-6)$$

where ϕ_o is the flux in the center of the slab. Notice ϕ is a maximum in the center ($x = 0$) and goes to zero when $x = \pm(a/2 + \delta)$. Generally, the flux goes to zero at an additional distance δ , called the extrapolation distance.

² It is clear that equation 3-4 can be written as $B_g^2 = -\nabla^2 \phi / \phi$. The quantity on the right is the curvature or buckling in mathematics, hence, the reason John Wheeler, a developer of fission theory and reactors, gave it the name buckling.

Fig. 3-1 Slab Flux Profile

Table 3-1 includes a factor δ in addition to the physical length to account for the extrapolation length or the sum of the extrapolation length and the reflector savings.

Figure 3-1 illustrates the fact that the geometric buckling is the distance from the peak flux to the extrapolated vanishing point. This is a very simplified exposition of buckling to illustrate the concept. The buckling of a complex reactor such as a process system criticality accident is much more complex.

Buckling in Regular Geometries

An infinite slab geometry is not realistic. Table 3-1 provides the geometric bucklings for a sphere, and a cube.

Table 3-1 Bucklings for Regular Geometries	
Geometry	Buckling B_g^2
Sphere, radius r	$[\pi/(r+\delta)]^2$
Cylinder, radius r , height h	$[2.405/(r+\delta)]^2 + [\pi/(h+2*\delta)]^2$
Cube, dimensions a, b, c	$[\pi/(a+2*\delta)]^2 + [\pi/(b+2*\delta)]^2 + [\pi/(c+2*\delta)]^2$

Critical Condition

The reactor becomes critical when the geometric and material bucklings are equal:

$$B^2 = B_g^2 = B_m^2, \quad (3-7)$$

therefore, the task of estimating criticality conditions is one of estimating the material and geometric bucklings. At critical, the two bucklings are the same; hence, they are designated by B^2 .

A reactor is subcritical when:

$$B_g^2 < B_m^2, \quad (3-8)$$

and supercritical when:

$$B_g^2 > B_m^2. \quad (3-9)$$

Tabulated M and k_{∞}

Extensive tabulations³ of M and k_{∞} have been prepared adjusted to make equation 3-6 correctly predict the critical condition.

Using these data, the k-factor can be calculated from the six-factor formula as:

$$k = k_{\infty}/(1 + M^2 B^2) \quad (3-10)$$

which gives a measure of the degree of subcriticality.

Effect of Density

A low density reactor is more likely to leak neutrons than a high-density reactor. The effect of density on the amount of mass necessary for criticality⁴ is:

$$m_{\text{crit}} \propto 1/\rho^2 \quad (3-11)$$

Equation 3-11 shows the importance of the high-explosive system that compresses an atomic bomb to make the reaction supercritical. By the same token, mass needed for criticality increases in proportion to the square of the density reduction.

Review Questions

1. Name and explain the terms in the six-factor formula.
2. What conditions correspond to: $k=1$, $k<1$, $k>1$?
3. Explain geometric buckling.
4. How does density affect the amount of fissionable material necessary for criticality.
5. What is the relationship between k , k_{∞} , M, and B?

³ R.D. Carter, G.R. Kiel and K.R. Ridgway, "Criticality Handbook", ARH-600, Volume III, 1980.

⁴ H.C. Paxton, "Criticality Control in Operations with Fissile Materials," LA-3366, Los Alamos Scientific Laboratory, 1972.

3.2 Measurement of Criticality

Theories alone, especially those formulated before modern computers, lacked sufficient precision for risking health and safety on the calculations. These theories were supported by experimental measurement of the conditions for criticality. In fact, experimental results were used to calibrate calculations and provide a basis for their refinements.

Critical Facilities

A critical facility is designed for determining the conditions for criticality. Because of the dangers involved, these facilities are remote and can be operated from a safe distance. Criticality measurements were performed before the Los Alamos Pajarito Site began operation in 1948, but this oldest of criticality facilities has regularly performed measurements on all three of the fissile species.

Criticality facilities exist or existed at most of the National Laboratories, and at some of the industrial reactor designers and nuclear material processors but usually focus on criticality determinations for their mission geometries and compositions. For example Argonne National Laboratory performed Zero-Power Reactor and Zero-Power Plutonium Reactor (ZPPR) experiments in support of the design of the breeder reactor.

Mechanically Separable Facilities

Experiments performed with solid fuels and moderators use mechanical methods to stop the reaction before damage is done. For a large assembly such as a reactor lattice, this takes the form of a horizontal or vertical split bed. The assembly is divided in half; one half is stationary and the other is movable. The two halves are slowly moved together, and neutron multiplication is measured. If multiplication becomes excessive, during the approach of the two halves, indicating the onset of criticality, the halves are quickly separated.

The process is similar for highly enriched, spherical geometries, such as those investigated at Los Alamos or Livermore Laboratories. The two halves, which may be bare spheres of uranium or plutonium can move together to determine the neutron multiplication and rapidly separated. In some experiments, super-critical assemblies are brought together to separate themselves by the large energy release from the chain reaction.

Liquid Dump Facilities

Experiments with solutions containing fissile materials are performed similarly. The solutions, contained in geometrically safe containers and pipes are slowly transferred into a container with the shape being investigated. If multiplication becomes excessive, valves rapidly open to discharge the solution into a safe geometry.

From many critical experiments, a criticality database has been constructed for special and standard geometries, combinations of fuel, moderator amounts, and types.

Measurement of Multiplication

Suppose we wish to measure the k-factor of a potentially critical reactor e.g. a spherical storage vessel which contains plutonium at different concentrations. The object of the measurements is to determine the critical concentration.

A neutron source is located in the vessel and one or more neutron detectors, such as boron-trifluoride counters, are located about the container. The counting rate of the neutron detectors are measured when the solution contains no plutonium; call this $R(0)$.

Adding plutonium increases k, and multiplies the number of neutrons in the reactor, resulting in a detected neutron counting rate $R(k)$, and a neutron multiplication of:

$$M = R(k)/R(0) \quad (3-12)$$

The neutron multiplication is:

$$M = R(0)*k + R(0)*k^2 + R(0)*k^3 + \dots /R(0) \quad (3-13)$$

where the definition of the k-factor is:

$$k = \frac{\text{number of neutrons in one generation.}}{\text{number of neutrons in the previous generation}}$$

Equation 3-13 is an infinite series that can be written as:

$$M = 1/(1-k). \quad (3-14)$$

and

$$k = (M-1)/M \quad (3-15)$$

Numerical Example

What is the k-factor of an unreflected sphere 10 cm radius which gives a neutron counting rate of 100 cpm when filled with a solution without plutonium, and 10,000 cpm when the plutonium concentration is 5 kg/l? The multiplication is 100. Substituting into equation 3-15: $k = 99/100 = 0.99$.

Extrapolated Criticality

Plotting the k-factors obtained from many neutron multiplication experiments as a function of a reactor parameter (such as plutonium concentration) enables the extrapolation of the

point at which the parameter would result in criticality. Graphs of parameters that result in criticality are prepared from a multitude of experiments to provide guidance for safe plant operations.

Review Questions

1. Describe a critical facility for a solid assembly.
2. Describe a critical facility for solutions.
3. What is the neutron multiplication factor of process container with a k-factor of 0.95?

Table 3-2 Single Parameter Limits for Uniform Nitrate Aqueous Solutions of Fissile Nuclides			
Parameter	U-235	U-233	Pu-239
Fissile mass (kg)	0.78	0.55	0.48
Cylinder dia. (cm)	14.4	11.7	15.4
Slab thickness (cm)	4.9	3.1	5.5
Solution volume (ℓ)	6.2	3.6	7.3
Fissile concentration (g/ℓ)	11.6	10.8	7.3
Areal density (g/cm ²)	0.4	0.35	0.25
Hydrogen/fissile atomic ratio	3250	2390	3630

Table 3-3 Single Parameter Limits for Metal Assemblies			
Parameter	U-235	U-233	Pu-238
Fissile mass (kg)	20.1	6.0	5.0
Cylinder dia. (cm)	7.3	4.5	4.4
Slab thickness (cm)	1.3	0.38	0.65
Enrichment	5%		
Maximum density	18.81	18.65	19.52

3.3 Subcritical Limits

Every process situation or measurement that may be encountered cannot be immediately evaluated experimentally or theoretically to determine if it is dangerous. For practicality, guidelines containing safety margins must be available. Single-parameter limits are the most restrictive; multi-parameter limits are less restrictive and allow a closer approach to the critical condition.

Single Parameter Limits

Table 3-2 provides subcritical limits for uniform aqueous solutions of the three fissile nuclides in which one single parameter, fissile mass, is controlled to prevent criticality. As long as the controlling parameter is not exceeded, criticality is not possible. The values assume that there is unlimited water reflection. The linear limits refers to the diameter of the infinite cylinder, or to the thickness of infinite slab. The limit on atomic ratio is equivalent to the limit on solution's concentration, but may be applied to non-aqueous solutions regardless of chemical composition.

Table 3-3 presents the single-parameter limits on fully water-reflected metal assemblies. The mass limits assume that there are no re-entrant holes filled with moderator.

Multiple-Parameter Limits

Conformance with the single-parameter limits would prevent the use of the 100,000 kg inventories of UO_2 that must be fabricated for an initial reactor-core loading. Multi-parameter limits relax the single-parameter restrictions through administrative constraints on operations. Control of solution concentration for specified geometries allows the use of multi-parameter limits.

Figure 3-2 shows the multiparameter limits for a spherical aqueous solution of plutonium. The ordinate is the critical mass and the abscissa is the critical radius. The upper curve presents data for a bare (unreflected) spherical container of the solution; the lower curve presents data for a fully water-reflected sphere. The lines at 45° angles indicate the

Figure 3-2 Critical Mass and Radius for a Bare and a Reflected Plutonium-Solution Sphere

plutonium concentration at the intersection of these lines with the curves. These data show that a fully reflected plutonium metal sphere, as small as 4 cm radius, density 19.6 g/cm₃, is critical if fully reflected and would contain 5.3 kg of plutonium. If the sphere is bare, a larger quantity, 9.8 kg, of plutonium is needed for criticality in which case the radius is 5 cm.

Figure 3-3 Multi-Parameter U-235 Spherical Sub-Criticality Limits

Figure 3-3 presents multiparameter limits for a sphere of U-235 aqueous solution as a function of uranium concentration and sphere mass. The lower curve applies to 30 cm of water reflection; the upper curve applies to 2.5 cm of water reflection. It is seen that the minimum uranium mass to go critical is 0.6 kg in a solution 0.05 kg U/l, or about 500 hydrogen atoms for every uranium atom.

Figure 3-4 shows multiparameter limits for U-235 solution in slab geometry. The left portion of the curve applies to a uranyl dioxide difluoride solution and the right to uranium metal mixed with water. The upper curve is for 2.5 cm thick moderation; the lower curve is for 30 cm thick moderation. It may be noted that slabs as thin as 1.3 cm are critical if fully reflected and in concentrations as high as 2 kg/l.

Fig. 3-4 Multi-Parameter U-235 Slab Sub-Criticality Limits

Review Questions

1. What is the purpose of criticality limits?
2. What is the single parameter limit for spherical fissile mass in nitrate aqueous solution mass?
3. What is the single parameter limit for spherical fissile mass in a metal assembly?
In a solution mass?
4. Why are multiparameter limits needed?
5. What is the smallest amount of plutonium that can be critical?
6. What is the smallest amount of U-235 that can be critical?
7. What is the radius of a fully reflected sphere of Pu-239 solution that will result in criticality?

3.4 Calculation of Criticality without a Computer

Unfortunately, there are many complex geometries and material combinations that occur in process and experimental situations than can be accurately represented by multiparameter criticality graphs. The criticality aspects of these complex geometries can be calculated with computers, but several methods have been developed that are simple enough for hand calculations.

Buckling/Shape Conversion

The buckling/shape conversion method determines the equivalent geometry for which criticality limits are available to the geometry being assessed for criticality by relating the buckling of one to the other. If the buckling is greater than the subcriticality limited

Fig. 3-5 Extrapolation Lengths for Cylinders Containing UO_2F_2
(h=height; d=diameter)

buckling shape, the unknown is deemed to be subcritical; it is deemed to be critical if it buckling is less.

However, to use these formulae, the extrapolation distance, δ , is needed. While the extrapolation distance is important, it need not be known precisely because it is usually small compared with the bucklings provided in Table 3-1 for several regular geometric shapes.

Figures 3-5 and 3-6 provide extrapolation length data for uranium and plutonium cylinders, specifically for uranium enriched to 93.2% in the isotope U-235 in Figure 3-5, while Figure

3-6 is for enrichment to 93.5%.

Fig. 3-6 Extrapolation Lengths for Cylinders of Uranium Metal and δ -phase Plutonium (h =height; d =diameter)

Example of Buckling/Shape Conversion

Suppose we have a fully reflected, cylindrical process container that is 30-cm high and 20-cm in diameter. The ratio $(h/d)/[1+(h/d)] = 1.5/(1+1.5) = 0.6$. Figure 3-5 gives the extrapolation length $\delta = 5.9$ cm. To find the spherical equivalent to the cylinder, we equate their bucklings from Table 3-1:

$$[\pi/(r+\delta)]^2 = [2.405/(r+\delta)]^2 + [\pi/(h+2*\delta)]^2 \quad (3-14)$$

or,

$$[\pi/(r+5.9)]^2 = [2.405/(10+5.9)]^2 + [\pi/(30+2*5.9)]^2, \quad (3-15)$$

where the same extrapolation length is used for the sphere as for the cylinder. The result is that the equivalent sphere is 12.7 cm radius, which is equivalent to a spherical volume of 8.58 ℓ . The concentration is $C = m/V$, where m is the mass and V is the volume. Substituting for V , $m = 8.58*C$. This is the equation of a straight line on a log-log plot with a slope of 45° which has been affixed to Figure 3-5, as shown in Figure 3-7. The intersection of this straight line with the fully reflected (lower) curve occurs at a mass of 7 kg of uranium which also corresponds with density of 0.815 kg/ ℓ . Thus, a concentration in excess of 0.815 kg/ ℓ will go critical. The volume of the cylinder is 9.724 ℓ , hence, the maximum subcritical uranium content is 7.68 kg of U-235.

In this example, we found the sphere equivalent to a cylinder. It also would have been correct to relate a finite cylinder to the criticality limits for an infinite cylinder. However shape conversion between quite different shapes, such as spheres and infinite slabs, may lead to error.

Surface Density Method

Fig. 3-7 Figure 3-3 Modified to Include Mass-Concentration Equation

Consider a fissile inventory storeroom stacked with shipping containers like the "birdcage"⁵ containers shown in Figure 3-8. If a catastrophe occurred and the material were deposited on the floor, forming a slab, and this slab became flooded with water, the configuration should still be subcritical. This requirement that an inventory must be subcritical even in flooded slab form is the basis for the surface density method. A well-verified formulation⁶

⁵ These containers are empty except for the small, centered container of fissile material. The open spacing increases the neutron leakage and provides safety from criticality.

⁶ J.T. Thomas, Ed., "Nuclear Safety Guide/TID-7016 Revision 2", NUREG/CR-0095, 1978.

is:

$$SD = 0.54 \cdot SD_o \cdot (1 - 1.37 \cdot f), \quad (3-16)$$

where SD_o is the surface density in gm/cm^2 of a critical water reflected slab, and f the mass of a unit in the array to the critical mass of an unreflected sphere of the same material ($f < 1/1.37$).

A storeroom stacked with such containers has a uniform spacing of material such that the surface floor density is:

$$SD = n \cdot m / d^2 \quad (3-17)$$

where n is the number of units stacked on each other, m is the mass of fissile material contained in each container, and d^2 is the length of a side of the square base of the shipping container.

Combining equation 3-16 and 3-17, the allowable center to center spacing for the storage of fissile materials is:

$$d = 1.37 \cdot \sqrt{\{n \cdot m / [SD_o \cdot (1 - 1.37 \cdot f)]\}} \quad (3-18)$$

Numerical Example

Consider fully enriched containers holding 5 kg each, stacked two high, forming a slab with a hydrogen-to-uranium ratio of 10. Figure 3-3 shows the mass of an unreflected sphere to be 15 kg, giving $f = 5/15 = 0.33$. The lower curve of Figure 3-4 gives a critical reflected slab thickness of 3.4 cm and a H:U ratio of 10, corresponding to a uranium concentration of 2.3 kg/l. From this, $SD_o = 2.3 \text{ g/cm}^3 \cdot 3.4 = 7.82 \text{ gm/cm}^2$. From equation 3-16, the allowed surface density is: $SD = 0.54 \cdot 7.82 \cdot (1 - 1.37 \cdot 0.333) = 2.3 \text{ g/cm}^2$. Using this in equation 3-17, the spacing is found to be: $d = \sqrt{(2 \cdot 5000 / 2.3)} = 65.9 \text{ cm}$. This is the closest safe spacing according to the surface density method.

Density Analog Method

In equation 3-11, we noted that the critical mass is inversely related to the square of the density:

$$m/m_o = (\rho/\rho_o)^2. \quad (3-19)$$

Fig. 3-8 Birdcage Shipping Container

Thus, if a critical mass, m_o having a density ρ_o , and the density of a reactor is known, the critical mass of the reactor may be calculated. A density relationship for an arbitrary array of N storage containers has been developed⁶:

$$N = [2.1 \cdot SD_o \cdot (1 - 1.37 \cdot f) / m]^3 \cdot V^2, \quad (3-20)$$

where the symbols have their previous meanings. For the special case of a cubic array, $V = d^3$, and $N = n^3$, where n is the number of containers stacked on top of each other. Solving equation 3-20 for d, the spacing:

$$d = \sqrt[n]{n \cdot m / [2.1 \cdot SD_o \cdot (1 - 1.37 \cdot f)]} \quad (3-21)$$

Numerical Example

Using the previous numerical example but let $N = 125$ containers ($n = 5$), then the spacing is: $d = \sqrt[5]{5 \cdot 5000 / [2.1 \cdot 7.82 \cdot (1 - 1.37 \cdot 0.33)]} = 52.9$ cm.

Contrasting the Surface Density and Density Analog Methods

The density analog method is not restricted to two tier stacking but it is restricted to N being the cube of an integer. The surface density method, as presented is restricted to two tier stacking but may be very large in the other dimensions. For an array of 20,000 containers, the density analog method gives a minimum spacing of 123 cm, while the surface density method still limits the spacing to 66 cm provided that administrative controls limits the stacking to two tiers.

Solid Angle Method

The last hand-calculational method to be discussed assumes there is no neutron absorption between the fissile containers. In such a case, equation 1-31 becomes equation 1-24 and the key aspect of the method is the calculation of the solid angle.

The solid angle method, when applied to an array of fissile packages refers to the maximum solid angle, Ω , if all of the packages have the same k, otherwise to the maximum product of $\Omega \cdot k$. The criterion expressed in the reference to footnote 6 is:

$$\Omega_{\max} = 9 - 10 \cdot k, \quad (3-22)$$

subject to the following restrictions:

- The k of any unit shall be less than 0.8,
- Each package shall be subcritical when submerged in water,
- The minimum surface-to-surface separation shall be 0.3 m,
- The maximum allowed solid angle shall be 6 steradians ($4 \cdot \pi$ steradians in a sphere).

Numerical Example of Solid Angle Calculation

Assume that the cylindrical fissile containers in a birdcage may be approximated as 7 cm radius spheres. Each container has $k = 0.8$; therefore, what is the closest spacing? Equation 3-22 says the maximum solid angle is 1 steradian. If packages for a regular array, there are 6 nearest-neighbor packages (assume the second nearest-neighbors are shadows by the first nearest-neighbors), hence the maximum solid angle is $1/6 = 0.167$ sr. The solid angle from a point to the surface of a sphere is: $\Omega = 2\pi\{1 - 1/\sqrt{1 + (r/h)^2}\}$, where r is the radius of the sphere and h is the minimum distance from the point to the surface of the sphere. Solving: $h = r[1 - \Omega/(2\pi)]/\{\sqrt{[\Omega/\pi]}*\sqrt{1 - \Omega/(4\pi)}\}$. Substituting values: $h = 7*[1 - 0.167/(2\pi)]/\{\sqrt{[0.167/\pi]}*\sqrt{1 - 0.167/(4\pi)}\} = 29.6$ cm. Since the spheres are 7 cm radius, we conclude the minimum center-to-center spacing is $14 + 29.6 = 43.6$ cm.

Review Questions

1. Explain the following methods for criticality control:
 - a) buckling/shape conversion,
 - b) surface density method,
 - c) density analog method,
 - d) solid angle method.
2. Why are birdcage shipping containers used for shipping and storage?
3. What is the extrapolation distance?
4. How could you increase the packing density of fissile material and still avoid criticality? Would your method be inherently safe or require administrative control?

3.5 Computer Calculation of Criticality

The 19th century saw parallel approaches to the theory of heat. Thermodynamics is based on relationships between observable quantities; statistical mechanics, as developed by Maxwell and Boltzmann, applies statistics to the classical mechanical descriptions of the motions and vibrations of atoms and molecules to determine average quantities. The original mathematics did not show that a non-equilibrium distribution would equilibrate to the Maxwell-Boltzmann which is the reason Boltzmann developed his transport equation:

$$[1/v(E)] * d\phi(\underline{r}, E, \underline{\Omega}, t) / dt = -\underline{\Omega} \cdot \nabla \phi(\underline{r}, E, \underline{\Omega}, t) - \Sigma_t(\underline{r}, E, \underline{\Omega}) * \phi(\underline{r}, E, \underline{\Omega}, t) + S(\underline{r}, E, \underline{\Omega}, t) + \int dE' * \int d\Omega' * [\Sigma_s(\underline{r}, E' \rightarrow E, \underline{\Omega}' \rightarrow \underline{\Omega}) * \phi(\underline{r}, E', \underline{\Omega}', t)], \quad (3-23)$$

where $\phi(\underline{r}, E, \underline{\Omega}, t)$ is the neutron flux at vector location $\underline{r}$, having energy E , going in the direction of the vector solid angle $\underline{\Omega}$, at time, t . $S(\underline{r}, E, \underline{\Omega}, t)$ is the strength of the source emitting neutrons, and $\Sigma_s(\underline{r}, E' \rightarrow E, \underline{\Omega}' \rightarrow \underline{\Omega})$ is the cross section for neutron scattering from $\underline{\Omega}' \rightarrow \underline{\Omega}$, changing energy from $E' \rightarrow E$ at location $\underline{r}$. This equation is called an integro-differential equation because it contains both integrals and differentials. Its only exact solution for neutron transport is for an infinite amount of hydrogen surrounding a point-isotropic neutron source.

Chain reactions are calculated by two basic methods: 1) approximate solution of the transport equation, and 2) simulation of the chain reaction process - called a Monte Carlo calculation.

Computer Approximations of the Transport Equation

Modern digital computers are not really mathematicians - they can only do arithmetic - much less solve integro-differential equations. A human must modify the transport equation into an arithmetic form and instruct the computer how to calculate the problem with instructions called the computer program.

This process consists of dividing the variables in the transport equation into small intervals over which the variable is assumed to be constant and equal to its interval average. For example, the energy may be considered to have 16 values covering the range from fission to thermal energy. Similarly, space comprised of the core, moderator, and reflector is divided into one, two or three dimensional spatial units. The angular dependence of the neutron flux may be divided in polar and azimuthal steps. After this division into parts is done, the transport equation is converted into a very large number of coupled algebraic equations which the computer can solve. Codes that perform such computations are:⁷include: DTF-IV⁸, DOT⁹, and TWOTRAN¹⁰. The last two are two-dimensional discrete

⁷ W.W. Engle, Jr., "A Users Manual for ANISN: A One Dimensional Discrete Ordinates Transport Code with Anisotropic Scattering," K-1693, Oak Ridge National Laboratory, 1967.

ordinates programs which require considerable computer capabilities or time. In addition their rectangular or cylindrical meshes limit the representation of irregular geometries. Errors may result from excessive mesh spacing which should be less than the neutron mean-free path length in the material.

Computer Simulation of the Chain Reaction: Monte Carlo

Computer simulation begins by assuming a neutron is emitted from a weighted randomly selected location in the source region; the weighting should correspond to the spatial emission distribution. The neutron is assigned a randomly selected direction that is weighted by the angular emission distribution. The neutron's energy is randomly selected from a distribution corresponding to the emission energy spectrum. It travels through various material corresponding to the problem for a distance randomly weighted by the transmission in the medium through which it is traveling. The type of interaction is selected randomly from the relative amounts of the competing cross-sections. If the neutron is captured and only neutron history is being calculated, this trial ends and the process is repeated by selecting another source neutron.

If the neutron scatters, the scattering angle and energy are calculated from the kinematics, and the neutron travels in this new direction at this new energy until it interacts in a distance randomly weighted by the transmission in the medium through which it is traveling. The type of process is selected as before, and this procedure continues until the neutron is captured. If it is captured in a fission process, the number of neutrons emitted are randomly selected from the fission neutron emission distribution and these neutrons must be traced. A neutron may come to the boundary of the reactor and be lost in space which also terminates a history. By repeating this neutron life-death process a very large number of times, and combining the trials, the neutron flux, $\phi(\underline{r}, E, \underline{\Omega}, t)$ is calculated. The process described is called an un-weighted Monte Carlo calculation. Its disadvantage is the very large number of trials needed to obtain an accurate answer. More sophisticated methods accelerate the convergence to give an accurate answer with fewer calculations.

Examples of Monte Carlo codes are KENO¹¹ and MONK¹².

⁸ K.D. Lathrop, "DTF-IV - A FORTRAN-IV Program for Solving Multigroup Equations with Anisotropic Scattering," LA-3373, Los Alamos Scientific laboratory, 1965.

⁹ R. Protsik, and E.G. Leff, "Users Manual for DOT2B: A Two-Dimensional Multigroup Discrete Ordinates Transport/Diffusion Code with Anisotropic Scattering," GEAP-13537, General Electric Co., 1969.

¹⁰ K.D. Lathrop, "TWOTRAN, A FORTRAN Program for Two Dimensional Transport " GA-8747, Gulf General Atomic Inc., 1968.

¹¹ J.T. West et al., "KENO-IV/CG, The Combinational Geometry Version of the KENO Monte Carlo Criticality Safety Program," NUREG/CR-0709, USNRC, 1979.

Cross Sections for Computer Calculations of Criticality

To run these complex calculations, a library of cross sections for all of the materials, all of the energies, and all of the processes are needed. This is a lengthy subject and only the criticality safety reference library will be mentioned¹³.

Review Questions

1. Describe a Monte Carlo calculation of a chain reaction.
2. Name four computer codes for calculating the neutron transport equation.
3. Name two Monte Carlo codes for criticality calculations.

Summary of Chapter 3

The chapter began by extending the four-factor formula by two additional terms to calculate non-leakage probabilities. Fermi's age diffusion theory was mentioned to show the relationship of the non-leakage probabilities to the material buckling, neutron age and migration length. The concept of geometric buckling was introduced, with formulae for simple geometries. It was stated that criticality occurs when the geometric and material bucklings are equal, noting that the critical mass is proportional to the inverse square of the density. The multiplication method for measuring criticality was presented with discussion of critical experiments. Single and Multiple criticality limits were presented, followed by four hand-calculational methods for estimating criticality - especially for storing fissile materials. The chapter ended with discussion of the computer calculation of criticality using the transport equation and by Monte Carlo simulation codes.

¹² D.E. Bendall, "MONK-6 - A Revised Method of the U.K. Criticality Code," Trans. Amer. Nucl. Soc. 41, 332, 1982.

¹³ W.E. Ford III et al., "CRSL-V: an ENDF/B-V 227 Group Cross Section Library for Criticality Safety Studies," Trans. Am. Nucl. Soc. 34, 331, 1980.

4. CRITICALITY ACCIDENTS¹ and THEIR PREVENTION

To this point, we have discussed the theory of criticality accidents; in this chapter, the reality is presented by a chronological discussion of major accidents, some of which were lethal.

4.1 Some Characteristics of Criticality Accidents

A criticality accident is like an explosion or a fire or both in the sense that considerable energy is released. This energy can melt, boil, and vaporize apparatus, boil liquids, blow apart structures, and produce hazardous missiles. Personnel can be killed or maimed by the energy release but the usual killer is radiation in the form of neutrons, and gammas. Betas and alphas can play a part, but their range is limited. To emphasize these points, a criticality accident may, depending on the severity:

- Throw personnel down and about,
- Rupture containers by steam pressure,
- Fracture equipment and produce flying missiles,
- Violently eject liquids,
- Melt, and possibly ignite, uranium and plutonium metals,
- Disperse radioactive particles,
- Ignite and explode surrounding materials,
- Emit lethal or incapacitating radiation.

Typically, a criticality accident produces an initial pulse of energy and radiation. The only protection for this is to be as far away from, and behind as much shielding material as possible. However, as will be seen from the following accident descriptions, many criticality accidents are of considerable duration. Immediately evacuating the area and prohibiting further entry significantly saves lives and personnel from radiation exposure. For evacuation to be successful, it is necessary to know that a criticality accident has occurred. In many of the following accidents, the instrumentation was faulty and it was not realized that a criticality accident had occurred until ozone was smelled or liquid was boiling. Had the instrumentation been proper or worked properly, the dose and destruction could have been greatly reduced. Another reason for accurate radiation measurements is to guide the modality of the medical treatment of exposed personnel.

The accident descriptions show that the Four Horsemen of criticality accidents are:

- Enrichment
- Mass
- Moderation
- Reflection

¹ This information is taken from: W.R. Stratton, revised by D.R. Smith, "A Review of Criticality Accidents," DOE/NCT-04, March 1989.

As we saw in the previous chapter, an amazingly small amount of highly enriched fissile material can go critical with moderation and reflection. The presence of enrichment should convey the message that it should be treated with caution. Further caution should be given when the mass of fissile material is significant. The last warning is the presence or potential presence of hydrogen-containing materials. Usually this material is water but it can take many forms with subtle ways of entry.

Finally, if a criticality occurs, it is essential to report and document it immediately, following all of the reporting procedures. The primary reason for reporting is to prevent this type of accident from recurring. The report will highlight any weaknesses in procedures, systems, or actions for correction.

The following accident descriptions were selected to show the diversity of how criticality accidents have occurred. Particular attention has been given to fatal accidents.

4.2 Selected Criticality Accidents

Un-moderated Critical Experiment, Los Alamos, August 21, 1945

This is the first recorded fatality from a criticality accident.

The accident involved two hemispheres of delta-phase plutonium coated with 0.005 inch of nickel partially reflected by tungsten carbide. The total core mass was 6.2 kg; the density was about 15.7 g/cm³. The experimenter was moving the final reflector brick over the assembly when the nearby neutron counters indicated that the addition of this brick would make the assembly supercritical. The brick slipped from his hand onto the assembly which went supercritical. His exposure, estimated as 510 rem from a yield of 1E16 fissions, caused death 28 days later. The nickel canning on the plutonium core did not rupture.

Moderated Metal Assembly, Los Alamos, June 6, 1945

Sphere-like water-reflected, enriched metal fuel was used in an experiment to determine the critical mass when surrounded by hydrogenous material. Uranium mass of 35.4 kg (average enrichment 79.2%) consisting of ½ inch cubes were stacked to approximate a sphere in a 6-in. polyethylene cube. The assembly, in a tank, became critical as the tank filled with water. No scram device was provided. The assembly was made subcritical in 5-10 seconds after a yield of 3-4E16 fissions that raised the temperature of the metal more than 200°C. The polyethylene box was not watertight and water may have entered the uranium assembly as the tank filled, thereby providing unexpected moderation.

Critical Experiment, Los Alamos, May 21, 1946 - Slotin Accident

This accident is remarkably similar to the accident described in section 4.2.

The system consisted of the same plutonium sphere but reflected by beryllium. The top hemisphere had one edge resting on the lower beryllium hemisphere with the opposite edge

separated by the tip of a screwdriver. The demonstrator was slowly removing the screwdriver when it slipped, and the gap between the hemispheres closed. A prompt criticality occurred with the top hemisphere being thrown to the floor; all personnel left the room; The yield of this excursion was $3\text{E}15$ fissions with no rupture of the nickel canning. The eight people in the room received doses of about 2100, 360, 250, 160, 110, 65, 47, and 37 rem. The demonstrator who received 2100 rem died 9 days later.

Water-Boiler Solution Reactor, Los Alamos, December, 1949

The water-boiler reactor, consisting of a 12 inch-diameter sphere containing 13.6 ℓ of a uranyl nitrate water solution reflected by graphite, was being used to test two new control rods. The operator was manually checking their drop-time. After several tests of each rod separately, both rods were pulled, held for about 5 seconds and dropped simultaneously; this test was repeated. The reactivity with both rods removed was about 3% over prompt critical and causing a reactivity excursion with a period of 0.16 s with a yield of $3\text{-}4\text{E}16$ fissions. The excursion was not immediately detected because all the instrumentation was turned off, except for a direct-reading thermometer that showed a rise of 25°C . The operator received 2.5 rem of gamma radiation, but the reactor was undamaged.

Y-12 Chemical Processing Plant, ORNL, June 16, 1958

The accident occurred in a processing area in which enriched uranium was recovered chemically from various materials in a complex of equipment. The recovery system was being remodeled, and the situation was further aggravated by an inventory in progress that required disassembly, cleaning, reassembly, and leak testing of certain pieces of equipment, particularly several long, safe geometry, 5 inch-diameter pipes used to store aqueous solutions of U-235. As a consequence of the overlapping operations and of the irregular operation of some valves, a quantity of enriched uranium solution was inadvertently transferred into a non-safe 55-gal drum which was critical for about 20 min. It is believed that the first spike was $\sim 1\text{E}16$ fissions for a total yield of $1.3\text{E}18$ fissions. The average power was ~ 220 kW. There was no damage or contamination, but eight people received radiation doses of: 461, 428, 413, 341, 298, 86.5, 86.5, and 28.8 rem. At least one person owes his life to the fact that prompt and orderly evacuation plans were followed. One person survived 14-1/2 years; one 17-1/2 years; the status of one is unknown; and five were alive 29 years after the accident.

Separated Phases in Plutonium Process Tank, LANL, December 30, 1958

The accident occurred while chemical steps were being taken to purify and concentrate plutonium from slag, crucible, and other lean residues in recovery processes. Typical solutions contained less than 0.1 g Pu/ ℓ . An annual physical inventory was in progress; thus, normal flow into the area was interrupted so that residual materials in all process vessels could be evaluated for their plutonium content. Unexpected plutonium-rich solids, which should have been handled separately, were washed from two vessels into a single large

vessel that contained dilute aqueous and organic solutions. After removing most of the aqueous solution from this vessel, the remaining approximately 200 ℓ of material, including nitric acid wash, was transferred to a 850-ℓ, 96-cm-diameter stainless steel tank where the accident occurred. The tank contained about 295 ℓ of a caustic-stabilized aqueous-organic emulsion; the added acid is believed to have separated the liquid phases. The bottom layer (330 ℓ) is thought to have contained 60 g Pu; the upper, organic layer (160 ℓ) contained 3.27 kg Pu. When the motor drive of a stirrer was started, the system went prompt critical. The yield was $1.5E17$ fissions. The accident resulted in the death, 36 h later, of the operator who was looking into a sight glass when the motor was turned on. His radiation dosage was estimated to have been 12,000 rem. Two other persons apparently suffered no ill effects after receiving radiation doses of 134 and 53 rem. No equipment was contaminated or damaged, even though the shock displaced the tank about 1 cm at its supports. The vessels were replaced with smaller, more geometrically safe vessels and improved techniques for the sampling of solids were implemented; the importance of adherence to procedural controls was emphasized.

Plutonium Recovery, Hanford Works, Richland, WA, April 7, 1962

This Recuplex process accident occurred during cleaning the floor of a solvent extraction hood, involved a product-receiver tank that could overflow into the hood, a temporary line running from the hood floor to a transfer tank, and the apparent improper operation of valves. The receiver tank overflowed into the hood, leaving a solution containing about 45 g Pu/ℓ on the floor and in the sump; contrary to standing orders, the operator opened the valve allowing the solution to be lifted to the transfer tank; the later addition of aqueous (moderating) solution (10-30 ℓ at 0.118 g Pu/ℓ following mixing, and de-aeration of the contents of the transfer tank led to the excursion. The yield was $8E17$ fissions, with an initial power spike of $1E16$ fissions, but the tank remained critical for 37.5 hours as the power steadily decreased. Activation of the building criticality alarm resulted in prompt evacuation. At the time, 22 people were in the building with 3 receiving exposures of 110, 43, and 19 rem. The incident caused no damage or contamination but did precipitate final shutdown of the plant.

Wood River Junction, Scrap Recovery, Rhode Island, July 24, 1964

This plant was designed to recover highly enriched uranium from unirradiated scrap material left from the fabrication of reactor fuel elements. Typical of the difficulties that should be expected with a new operation, an unexpectedly large amount of uranium-contaminated trichloroethane (TCE) solution had accumulated. The very low concentration of uranium in the solution was recovered by mixing the TCE with sodium carbonate solution. Before July 17, 1964, the operation was performed by hand in small geometrically safe bottles. Because of the large amount of solution, the operation was shifted to a geometrically unsafe sodium carbonate makeup tank approximately 18 in. in diameter and 25 in. On the day before the accident, a plant evaporator had failed to operate properly and a plug of uranium nitrate crystals was found in a connecting line. The crystals were

dissolved with steam, and the resulting concentrated solution (240 g $^{235}\text{U}/\ell$) was drained into polyethylene bottles identical to those that normally held the very low concentration TCE solution. A bottle of the concentrated solution was mistaken for the TCE solution, and the operator poured it into a makeup tank that contained 41 ℓ of sodium carbonate solution being agitated by an electric stirrer; this caused criticality. The excursion ($1.0\text{--}1.1\text{E}17$ fissions) created a flash of light, splashed 20% of the solution out of the makeup tank, and knocked the operator to the floor. He regained his footing and ran from the area to an emergency building some 200 yards distant. The radiation dose of 10,000 rem to which he was exposed caused death 49 hours later. An hour and a half after the excursion, two men entered the area to drain the solution into safe containers. They turned off the stirrer, causing a change in geometry causing a second excursion. The estimated yield of the second excursion was $2\text{--}3\text{E}16$ fissions, but it was not realized that there had been a second excursion until much later because the alarm was still sounding for the first event. The two men involved in the second excursion received radiation doses, apparently while they were departing, estimated at between 60 and 100 rem. Other persons in the plant received very minor doses. No physical damage was done to the system, although the splashed solution had to be cleaned up. The total yield was $1.3\text{E}17$ fissions.

Idaho Chemical Processing Plant, Oct. 16, 1959

This chemical processing plant accepts fuel elements from various reactors, especially naval reactors. The active material (34 kg of enriched uranium, 93% U-235 in the form of uranyl nitrate concentrated to about 170 g $\text{U-235}/\ell$) was stored in a bank of containers with favorable geometry. The inadvertent initiation of siphoning during an air-sparging operation resulted in the transfer of about 200 ℓ of the solution to a 5000-gal tank containing about 600 ℓ of water. The excursion yielded $4\text{E}19$ fissions, or sufficient heat to boil away nearly half of the 800 ℓ solution. After the incident, much of the uranyl nitrate was found to be crystallized on the inner walls of the tank. Because of thick shielding, none of the personnel received significant gamma or neutron doses. During evacuation of the building, airborne fission products resulted in beta dosages of 50 R (one person), 32 R (one person), and smaller amounts to 17 persons. Equipment was not damaged.

Idaho Chemical Processing Plant, Jan. 25, 1961

This incident resulted from a bubble of high-pressure air (from an earlier operation unplugging a line) forcing about 40 ℓ of 200 g $\text{U-235}/\ell$ uranyl nitrate solution up a 5 inch-diameter pipe into a 2 ft. diameter, 4 ft. high vapor-disengagement cylinder. The excursion occurred in the cylinder, as a fast transient criticality. The yield was $6\text{E}17$ fissions. The part of the plant involved in the accident had been idle for about 12 months before the run.

Idaho Chemical Processing Plant, Oct. 17, 1978

The accident occurred in a shielded cell of a fuel reprocessing plant in which solutions from the dissolution of irradiated reactor fuel were processed by PUREX solvent extraction to

remove fission products and recover the enriched uranium. In the process of solvent extraction, immiscible aqueous and organic streams counterflow with intimate contact and, through control of acidity, a material of interest can be transferred from one stream to the other. In normal operation of the first column (1A), uranium went with the organic phase and fission products with the aqueous phase, but a small amount of uranium was carried by the aqueous - a concentration of about 0.15 g U/ℓ, so the aqueous stream from the second column (1B) was fed back and blended with the dissolver product going to 1A. The organic product stream from 1B, normally about 0.9 g U/ℓ, went on to the third column (1C), where the uranium was stripped from the organic by 0.005M nitric acid. The output of the stripping column went to mixer settlers where additional purification took place. Still further downstream, the uranium solution went to an evaporator where it was concentrated to permit efficient recovery of the uranium. Several factors contributed to this accident. An evaporator had plugged, and operations had been suspended for several weeks while the difficulties with instrumentation were corrected. During the downtime, a valve leaked water into the aluminum nitrate makeup tank used to prepare the aqueous feed to 1B. Over time, this leak diluted the feed solution from 0.75M to 0.08M. The 13,400-ℓ makeup tank was equipped with a density gauge that would have indicated the discrepancy, but the gauge was inoperable. The out-of-specification aqueous feed to the scrubbing column caused it to operate as a stripper rather than as a scrubber. Some of the enriched uranium was removed from 1B organic and recycled into the input of 1A. This partially-closed loop caused a steady increase in the uranium inventory in the two columns. Each time diluted solution was added to the feed tank from the makeup tank, the aluminum nitrate concentration in the feed was further reduced and stripping became more effective until the excursion occurred. A radiation alarm was activated by fission products in the plant stack's gasses. Soon after the alarm, several other alarms activated and the stack monitor gave a full-scale reading. The shift supervisor and the health physicist went outside the building and detected radiation intensities up to 100 mrem/h. The shift supervisor ordered the building evacuated; road blocks were established and management was notified. In the evacuation, the process operator shut off all feed to the first-cycle extraction process, but did not stop the pulsing of the columns. The reaction clearly took place in the lower section of 1B, with most of the fissions occurring in the upper portion of the column. The yield was 2.7E18, for an energy release of 165 MJ. The average power during the approximately 0.5 hour of the reaction was about 100 kW. There was no significant personnel exposure and no damage to process equipment.

As a direct result of this event, the plant suffered an extended and expensive shutdown; all operating procedures were reviewed in detail and revised. Increased emphasis was given to plant maintenance and to operator training. An extensive and highly instrumented plant-protection system was installed involving redundant sensors and redundant, automatic safety controls. The importance of maintenance of safety-related equipment and the need for adherence to well-developed operating procedures were reemphasized by this accident.

Vinca, Yugoslavia, Oct. 15, 1958

The critical facility at the Boris Kidrich Institute in Vinca, Yugoslavia, was a D20-moderated, unshielded, natural uranium. The total mass of uranium in the core was 3,995 kg in aluminum-clad fuel rods occupying a volume of $6.36\text{E}6\text{ cm}^3$. Two cadmium safety rods were installed but not interlocked with the flux recorder. The liquid level was normally used to control the system's reactivity (critical level, 178 cm). At the time of the accident a subcritical foil-counting experiment was in progress. To activate the foils as much as possible, it was decided to raise the multiplication to some high, but still subcritical, level. This was done by raising the heavy water in the tank in a series of steps. On the last step, two of the BF_3 chambers performed as before - leveling off at a higher signal level, but the third behaved erratically and was disconnected. After the assembly had been at this D20 level about 5 to 8 min, one of the experimenters smelled ozone and realized that the system was supercritical at some unknown power level. The cadmium safety rods were used to stop the reaction. Later investigation showed that both of the detector that were believed to be working properly had reached saturation and were reading a constant maximum value even though the power level was rising steadily. Six people suffered intense radiations estimated at 205, 320, 410, 415, 422, and 433 rem. One person died and the other five recovered after severe cases of radiation sickness. The yield was 80 MJ or $2.6\text{E}18$ fissions.

Stationary Low-Power Reactor No. 1, Idaho Reactor Testing Area, Jan. 3, 1961

The SL-1 was a direct-cycle, boiling-water reactor of 3 MW gross thermal power using enriched uranium fuel plates clad in aluminum, moderated, and cooled by water. Because the reactor was designed to operate for 3 years with little attention, the core was loaded with excess U-235; to counterbalance this, a burnable poison (B-10) was added. Because the boron plates had a tendency to bow and increase the reactivity, some of them were replaced in November 1960 with cadmium strips welded between thin aluminum plates. At that time, the shutdown margin was estimated to be 3% compared to the initial value of 3.5-4%. The cruciform control rods were large cadmium sheets sandwiched between aluminum plates. The poor condition of the core may have been irrelevant to the accident. After having been in operation for about 2 years, the SL-1 was shutdown for routine maintenance; it was to be brought to power again. The three-man crew on duty the night of the accident was reassembling the control rod drives and preparing the reactor for startup. The evidence suggests that the central rod was manually pulled out as rapidly as the operator was able to do so. This placed the reactor on about a 4-ms period; power continued to rise until thermal expansion and formation of a steam void quenched the excursion. The peak power was about $2\text{E}4$ MW, and the total energy release was 133 MJ. The subsequent steam explosion destroyed the reactor and killed two men instantly; the third died 2 hours later from a head injury. The reactor building, and, especially, the reactor room were very seriously contaminated by the reactor water, which carried fission products with it. Initial investigations were hindered by the high radiation field (500 to 1000 R/h) in the reactor room. In spite of the large radioactivity release from the core, very little escaped from the building, although it was not designed to be airtight.

RA-2 Facility, Buenos Aires, Argentina, Sept. 23, 1983

Control rods for this essentially zero-power experimental reactor facility were MTR (National Reactor Test Station, Materials Test Reactor - MTR) elements in which 4 of the 19 fuel plates were removed and replaced by 2 cadmium plates. Just outside the fueled region (approximately 30.5 cm x 38 cm) was a graphite reflector approximately 7.5 cm thick. The large reactor vessel was filled with demineralized water during operations and was supposed to be drained during changes in fuel configurations when people were required to be present. The technician, a qualified operator with 14 year's experience, was alone in the reactor room changing in the fuel configuration. The moderator had not been drained from the tank, though required by procedures. Two fuel elements had been placed just outside the graphite, instead of being removed completely from the tank. Two of the control elements, without the cadmium plates installed, were being placed in the fuel configuration. Criticality occurred as the second of these control elements was being installed. The excursion consisted of $3-4.5 \times 10^{17}$ fissions; the operator received about 2000 rad gamma and 1700 rad neutron, and lived for 2 days. Twelve other people received exposures: three received 20, 15, 1, and nine people received less than 1 rad.

Chernobyl Nuclear Power Station, Ukraine, April 26, 1986

Four RBMK-1000 (1000 MWe) graphite-moderated, water-cooled reactors comprises this power station. Unit 4 was the newest of these 12 m diameter, 7 m graphite core reactors operating on 2% enriched uranium. Because of the large spacing in the reactor coolant tubes, the void coefficient is positive. The operators were conducting an experiment to determine how long the turbine generator could provide power following scram. The reactor was at low power when the operators scrambled the reactor and halved the feedwater flow. Reduction of flow resulted in water boiling thus increasing reactivity because of the positive void coefficient. The process was regenerative - increasing the power a hundred-fold. The transient was terminated by a steam explosion that destroyed part of the core, reactor building, and releasing fission products to the environment. Fire accompanied the accident, releasing fission products. By late August 1986, 31 people had died from the accident.

Chemical Explosion at Kyshtym, Southern Urals, USSR, September 29, 1957

This explosion was reported as a chemical accident but is included here because of the large radiological effects.

Kyshtym was a nuclear weapons facility concerned with recovery of plutonium with consequential waste production. The facility started about 1949 using the sodium acetate process. Three large 80,000 gal. waste storage tanks were involved holding 2×10^7 Ci of waste in 100 gm/l concentration with sodium acetate in 60-80 gm/l concentration. The cooling on one tank failed and the instrumentation was faulty. Water in the solution evaporated, the temperature continued to rise, and at 380°C, a nitrate explosion occurred with about 10

tons of TNT yield that lifted the 160 ton concrete shielding cover from the tank, releasing 2 million Ci (Chernobyl released 50 million Ci). The two other tanks were damaged. An evacuation was ordered of the 23,000 km² area about the facility involving 10,000 people. Before the accident, the operators fled from the heat emanating from the tank, which may be the reason that no fatalities were reported.

4.3 Summary Tables

Tables 4-1 through 4-4 summarize 41 criticality accidents of fission yields from 1E16 to 1.2E20 with most accidents toward the low end of the range. The earliest accident was 1945 and the latest was in 1986. Table 4-1 through 4-4, respectively, list chemical process accidents, bare and reflected system accidents, moderated metal and oxide system accidents, fissile solutions, and miscellaneous accidents. For more details on the accidents in these tables see the reference in footnote 1.

Review Questions

1. How many criticality accidents have resulted in fatalities?
2. What accident produced the highest fission yield?
3. What was the TNT equivalent of the accident described in section 4.9?

Summary of Chapter 4

This chapter began with a discussion of the manifestations of a criticality accident as heat, explosion, and lethal radiation. Distance, shielding, prompt detection, mitigation, and evacuation are ways to reduce the consequences. This overview was followed by case histories to illustrate these points. The chapter ends with tables of 41 historical criticality occurrences - the worst and most recent was the Chernobyl accident. All in all, the frequency of criticality accidents has been diminishing with time. Reasons for the current rarity may be due to:

- Dangers from nuclear criticality are understood from experience that has been gained over the years,
- Nuclear research has advanced from the initial experimental stage,
- Criticality calculations can be performed very accurately,
- Instrumentation is excellent,
- Written procedures are required,
- Peer review of proposed experiment or process is required, and

- **Standards, Guides and Orders specify allowable actions.**

In hindsight, all of the accidents discussed in this chapter were preventable.

Table 4-1 Process Accidents						
Date	Location	Material	Geometry	Cause	Physical damage	Total fissions ²
6/16/58	Y-12, ORNL	2.5 kg U-235 nitrate	55-gal. drum	Wash water added to U-235 solution	none	~1E16
12/30/58	Plutonium recovery, LANL	3.27 kg Pu in 2 phase systems	250 gal. cylindrical tank	Stirrer changed geometry to super-critical	none	1.5E17
10/16/59	ICPP, Idaho	34.5 kg U-235 in ~800 ℓ water	5000 gal. cylindrical tank	Solution inadvertently siphoned	none	~1E17
1/25/61	ICPP, Idaho	8 kg U-235 in 40 ℓ water	Cylindrical disengagement section	Solution moved to non-safe geometry	none	~6E17
4/7/62	Recuplex plant, Hanford WA	1.55 kg Pu	Cylinder	Vacuum transfer of rich solution to large tank	none	~1E16
7/24/54	Wood River junction, RI	2.64 kg U-235	cylinder	solution poured into non-safe tank	none, 1 death	~1E17
8/24/70	BNFL Pu plant, Windscale, England	2.15 kg Pu	cylinder	Pu stripped from aqueous to trapped solvent	none	~1E15
10/17/78	ICPP, Idaho	8.45-10.55 kg 89% U-235, 7.61-9.31 kg U-235	Cylindrical scrub column	U-235 stripped from solvent by non-specification aqueous stream	none	~3E18

² One pound of TNT has the explosive yield of 7.25E16 fissions.

Table 4-2 Bare and Reflected Metal System Accidents						
Date	Location	Material	Geometry	Cause	Physical damage	Total fissions
8/21/45	LANL, NM	6.3 kg Pu	Sphere with tungsten carbide reflector	Hand stacking of reflector	none, 1 death	$\sim 1\text{E}16$
4/21/46	LANL, NM	6.3 kg Pu	Sphere with Be reflector	Hand control of sphere	none, 1 death	$\sim 3\text{E}15$
3/1/51	LANL, NM	62.9 kg 93% U-235	Cylinder with annulus in water	Inappropriate scram design	Minor damage to parts	$\sim 1\text{E}17$
4/18/52	LANL, NM	92.4 kg 93% U-235	Cylinder unreflected	Computation error	None	$1.5\text{E}16$
2/03/54	LANL, NM	53 kg 93% U-235	Sphere unreflected	Incorrect operation	Slight damage to parts	$5.6\text{E}16$
2/12/57	LANL, NM	54 kg 93% U-235	Sphere unreflected	Shifting of experiment	Severe damage to assembly	$1.2\text{E}17$
6/17/60	LANL, NM	~ 51 kg 93% U-235	Cylinder with 9 in. reflector	Excess material added	Slight	$6\text{E}16$
11/10/61	ORNL, TN	~ 75 kg 93% U-235	Paraffin reflected	Excess material added	None	$\sim 1\text{E}16$
3/26/63	LLNL, CA	47 kg 93% U-235	Cylinder with Be reflector	Held part fell	Gross damage to assembly	$3.7\text{E}17$
5/28/65	White Sands, NM	96 kg 93% U-235	Cylinder unreflected	Incorrect operation	Bolts broken, minor damage	$1.5\text{E}17$
9/6/68	Aberdeen PG, MD	123 kg 93% U-235 Mo alloy	Cylinder unreflected	Incorrect operation	Gross damage to assembly	$6.1\text{E}17$

Table 4-3 Moderated Metal and Oxide System Accidents						
Date	Location	Material	Geometry	Cause	Physical damage	Total fissions
6/6/45	LANL, NM	35.4 kg 79.2% U-235 cubes	Water reflected psuedo sphere	Water leaked into assembly	Minor	~4E16
~ 1950	Chalk River, Ont. Canada	Aluminum clad, natural uranium	Rods in heavy water moderator	Excess moderator added, unmonitored	Minor	?
6/2/52	ANL, ID	93% enriched UO ₂ in plastic	Fuel elements in water moderator	Control removed, water not drained	Extensive to fuel elements	1.2E17
12/12/52	Chalk River, Ont. Canada	Natural uranium fuel rods	Heavy-water moderated reactor	Positive void coefficient	Extensive to core and supports	1.2E20
7/22/54	Reactor test station, ID	4.16 kg 93% U-235 as U/Al alloy	Fuel elements in water moderator	Planned transient exceeded	Extensive	4.7E18
10/15/58	Vinca, Yugoslavia	Natural uranium rods	Fuel rods in heavy water	Faulty power monitoring	None, 1 death	~2.6E18
3/15/60	Saclay, France	2.2 tons 1.5% U-235 as UO ₂	Fuel rods in water	Removal of poison rod	None	3E18
1/3/61	Reactor test Station, SL-1, ID	93% U-235 metal clad in aluminum	Fuel rods in water	Removal of control rod	Extensive, 3 deaths	4.4E18
11/5/62	Reactor test station, ID	93% U-235 Al alloy plates, Al clad	Fuel elements in water	Planned transient exceeded	Extensive	~1E18
12/30/65	Mol, Belgium	7% U-235 UO ₂	Rods in water/heavy water	Misoperation and undrained tank	None	~4E17
9/23/83	Buenos Aires, Argentina	Materials Test Reactor-type fuel	Pool type reactor	Failure to drain tank	None, 1 death	~4E17

Table 4-4 Fissile Solutions and Miscellaneous System Accidents						
Date	Location	Material	Geometry	Cause	Physical damage	Total fissions
12/49	LANL, NM	~ 1kg U-235 as uranyl nitrate	sphere, graphite reflected	Withdrawal of two control rods	None	~ 3E16
11/16/57	Hanford, WA	1.15 kg PuNO ₃	Bare sphere 93 % filled	Control rods withdrawn too rapidly	None	8E16
5/26/54	ORNL, TN	18.83 kg U-235 UF ₆	Cylindrical annulus, bare	Tilting of inner cylinder	None	1E17
2/1/56	ORNL, TN	27.7 kg U-235 UF ₆	Cylinder, bare	Scram plate changed geometry	Slight	1.6E17
1/30/68	ORNL, TN	0.95 kg U-235	Sphere, water reflected	Air bubble added solution to sphere	Local contamination	1.1E16
2/11/45	LANL, NM	UH in styrex	The Dragon assembly	Yields increased until damage	Blistering of plastic	~ 4E17
~ 1954	USSR	Unknown	Unknown	Unknown	Unknown	~ 1E16
11/29/55	ANL, ID	Enriched uranium in NaK	EBR-1	Delayed scram during transient	Extensive core melting	~ 4E17
7/3/56	LANL, NM	93% U-235 foils in graphite	Honeycomb	Rapid reactivity addition	None	3.2E16
11/18/58	Reactor test station, ID	UO ₂ in nickel-chromium	Aircraft engine prototype	Instrumentation failure	Some fuel melting	2.5E19
12/11/62	LANL, NM	Large 93% U-235 graphite cylinder	Cylinder plus annular reflector	Faulty start-up operations	None	~ 3E16
4/26/86	Chernobyl, Ukraine, USSR	Large graphite-moderated, water cooled reactor	RBMK-1000 type	Faulty procedures, positive void coefficient	Severe, fire, 31 deaths	5E7 curies

5. STANDARDS, GUIDES, AND ORDERS¹

Chapters 1 through 3 of this course provide a background understanding of the physics of criticality. Not everyone can or desires to apply first principles to their work. Fortunately, rules based on theory, experience, and practice have been established for prudent guidance. Safety can be assured by conformance with the rules provided in the form of standards, guides, and orders. Standards are voluntary, but you violate them at your own risk. In that sense, they are not voluntary. Additionally, many of the standards are endorsed in guides and orders, thus attaining a level of legal requirement. Guides, used here is refers to the body of USNRC laws, executive orders, and Nuclear Regulatory Guides. These are compulsory for industry but not necessarily for DOE operations; however, recently there has been a tendency for DOE orders to endorse and extend NRC requirements, as well as to promulgate requirements not addressed by the NRC.

The message is that standards, guides and orders are to help you not hinder you. It is much easier to comply than to seek a variance which may be needed for a special purpose.

5.1 ANSI/ANS Standards

In 1918, five US engineering societies founded the American Engineering Standards Committee to establish material standards. Ten years later, three federal agencies joined and the organization was renamed the American Standards Association. The ASA grew to an organization of 100 societies and 2300 corporate members; this proved to be cumbersome. A US Department of Commerce study, in 1963, recommended changes and renaming to the United States of America Standards Institute (USASI) to serve the entire US economy. However, to avoid the appearance of being an arm of the Federal government, it was renamed American National Standards Institute (ANSI)². Within ANSI, is the Nuclear Standards Board, composed of 25 members including the American Nuclear Society that is the secretariat for N16, nuclear criticality safety.

Figure 5-1 shows the ring of criticality standards by their current designations. The general standard is ANS-8.1³ that interacts with the other 9 standards that interact with each other. Standard 8.3 addresses criticality alarms: 8.5; borosilicate glass absorbers in process lines: 8.6; safety in neutron multiplication experiments: 8.7; storage safety: 8.9; pipe intersections of fissile solutions: 8.10; safety control of shielding and confinement: 8.12; safety with homogenous fissile mixtures: 8.15; special actinide safety control: 8.17; safety in transporta-

¹ Much of the material for this chapter is taken from chapter 4 and 9 of R.A. Kneif, Nuclear Criticality Safety, American Nuclear Society, La Grange Park, IL, 1991.

² American Standard was already a copyrighted name of a plumbing manufacturer.

³ The correct designation is ANSI/ANS-8.1. The ANSI designation is not shown because ANSI is the overall standards organization.

Figure 5.1 The Ring of ANSI/ANS Standard Protection

tion: 8.19; administrative practices: and 8.20; nuclear criticality safety training.

ANS-8.1-1983 Nuclear Criticality Safety in Operations with Fissionable Materials Outside of Reactors and Validation of Calculational Methods for Nuclear Criticality Safety.

This general standard, among other things delineates administrative responsibility in criticality safety in defining:

- Safety responsibility and criteria,
- Process analysis, including the effects of credible abnormal conditions,
- Written procedures,
- Controlled movement of fissionable materials,
- Posting of material limits,
- Prompt reporting, investigation, and correction of processes and procedures affecting safety,
- Frequent independent operational reviews,
- Emergency procedures,
- Training.

Defense in Depth

The standard recognizes the difficulty, if not impossibility, of anticipating every possible occurrence in the conduct of an operation and requires a double contingency. No single mishap shall lead to criticality; at a minimum, two independent mishaps must occur before criticality can result.

For example, a fuel fabrication line is designed to exclude water, but the quantity of material and shapes of the powder-containing receptacles is such that they cannot be critical under full flooding.

In this example, safety is assured by: 1) moderator/reflector control, and 2) material quantity control. Other independent protection barriers are: 3) geometry control - flat geometries to increase leakage, and 4) absorber control - building absorbers into the equipment to prevent removal.

Subcritical Limits

The standard contains tables of single parameter limits for aqueous solutions of fissile material and for metal systems of the three isotopes.

Multi-parameter limits are provided as graphs, such as mass limits for uranium water lattices, cylindrical diameters for uranium-water lattices, slab thicknesses, volume limits, and areal density limits.

Safety Margins

The standard requires "adequate" safety margins but does not specify adequacy, which is left to the facility's management.

ANS-8.3-1979 Criticality Accident Alarm Systems

The purpose of this standard is to provide minimum alarm requirements for operations involving fissile materials in which inadvertent criticality may occur and cause personnel exposure in excess of the radiation protection guides published by the National Council on Radiation Protection (NCRP). The alarm shall be a distinctive sound capable of being heard in all parts of the area to be evacuated. The alarm trip point shall be set high enough to suppress spurious trips. Reliability may be provided by redundant channels; the design shall be such that testing will not cause an evacuation; it shall operate during a power outage. Accidents as short as $1\text{E-}3$ sec duration delivering greater than 25 rad 6 feet from the reacting material shall be detected. If the minimum accident cannot reach this dose level, alarms need not be provided.

Evacuation by the quickest routes shall be prepared and defined leading to personnel

assembly stations. Drills shall be performed at least annually and announced in advance. Emergency procedures coordinating with local and offsite organizations shall be prepared and approved by management.

Planning shall include dosimetry of affected individuals, environmental monitoring, accident diagnostics and medical arrangements for the care and treatment of injured and exposed personnel.

ANS-8.5-1979 Use of Borosilicate-Glass Raschig Rings as a Neutron Absorber in Solutions of Fissile Material

Borosilicate glass Raschig rings shall be used for neutron-absorption criticality control in chemical and radiological environments not exceeding the severity specified in the guide. The boron-10 to boron-11 ratio shall not be less than 24% and the glass shall contain 3.66-4.28 wt % boron. Provision shall be made for verifying the position and condition of the rings in the process equipment. The time between inspection shall not exceed 13 months, nor 7 months for agitated applications.

ANS-8.6-1983 Safety in Conducting Subcritical Neutron-Multiplication Measurements in Situ.

A written procedure for each new experiment shall be prepared and reviewed as required by management. It shall be reviewed by everyone taking part in the experiment. At least two persons shall be present during the experiment. Records emphasizing safety shall be kept during the experiment. Emergency procedures and radiation detection instrumentation shall be provided. The operation capability of the apparatus shall be verified before the experiment. The neutron source shall be of sufficient strength to provide a meaningful indication of multiplication. At least two neutron detectors shall be used. An audible indication of the neutron level shall be provided.

ANS-8.7-1982 Guide for Nuclear Criticality Safety in the Storage of Fissile Materials

Limits on the storage of fissile materials shall be based on experimental data or validated computational techniques. The facilities shall embody good engineering practices including consideration for fire, flood, earthquake and other calamities. Proper spacing of fissile materials shall be provided by physical barriers. The storage area should be maintained essentially free of combustible materials; a fire protection system must be included with drainage to preclude water accumulation. Criticality alarms shall be provided as appropriate.

The standard includes many table limiting the mass of uranium per cell in water-reflected arrays.

ANS-8.9-1978 Nuclear Criticality Safety Criteria for Steel-Pipe Intersections Containing Aqueous Solutions of Fissile Materials

This standard gives criteria and data based on criticality experiments and validated calculations which are applicable to homogenous aqueous solutions of fissile materials. The information is for solutions of highly enriched U-235, U-233, plutonium and uranyl solutions enriched <5 wt %. Four tables are provided for schedule 10 or heavier pipes.

ANS-8.10-1983 Criteria for Nuclear Criticality Safety Controls in Operations with Shielding and Confinement

Confined and shielded operations with the three fissile isotopes are discussed in this standard for protecting personnel, and the public outside the enclosure. All operations within the enclosure are conducted remotely by persons outside the shielding. The shield is adequate to meet radiation guidelines. The radiation dose for the maximum credible accident shall not exceed 25 rem nor exceed 0.5 rem for anyone outside of the restricted area. The structural shall not be damaged by credible accidents. Protection may be provided by distance in addition to the shielding.

ANS-8.12-1987 Nuclear Criticality Control and Safety of Plutonium-Uranium Fuel Mixtures Outside Reactors

Basic criteria are presented for plutonium-uranium mixtures in single units of simple shape containing no more than 30 wt % plutonium combined with uranium containing no more than 0.71 wt % U-235. The standard is for slurries of uniformly distributed particles with diameter less than 0.005 inches. The data are summarized in a table.

ANS-8.15-1981 Nuclear Criticality Control of Special Actinide Elements

ANS-8.1-1983 provides single parameter limits for the three "common" fissile isotopes. This standard extends single-parameter limits to rare fissile isotopes. Isotopes having an odd neutron number, such as Pu-241, Am-243, Cm-243, Cm-245, Cm-247, Cf-249, and Cf-251 may have critical masses less than 1 kg, and in some cases, much less. Those with an even number of neutrons, Np-237, Pu-238, Pu-240, Am-241, Am-243, and Cm-244 are threshold fissioners. Moderation causes them to become more subcritical. Subcritical mass limits are given for isolated assemblies. Multi-parameter limits are presented for quantities in excess of the single parameter mass limits. There are extensive tables and information about the isotopes.

ANS-8.17-1984 Criticality Safety Criteria for the Handling, Storage, and Transportation of LWR Outside Reactors

The potential for criticality accidents during the handling, storage, and transportation of fuel

for nuclear reactors presents a health and safety risk to the personnel involved and to the public. The standard endorses all other standards and specifies a neutron multiplication factor:

$$k_s \leq k_c - \Delta k_s - \Delta k_c - \Delta k_m \quad (5-1)$$

where, k_s is the calculated allowable maximum multiplication factor, k , of the system for all normal and credible abnormal conditions, k_c is the mean k that results from the calculation of the benchmark criticality experiments using a particular calculational method, Δk_s is an allowance for statistical or convergence uncertainties, Δk_c is a margin for uncertainty in the critical experiments, statistics, convergence, extrapolation of k_c , uncertainties in geometrical or material representations.

ANS-8.19-1984 Administrative Practices for Nuclear Criticality Safety

This guide specifies the responsibilities of:

- Management,
- Supervisors, and
- Criticality Safety Staff

These responsibilities concern:

- Operating procedures,
- Process Evaluation for Nuclear Criticality Safety,
- Materials Control, and
- Planned Response to Nuclear Criticality Accidents.

ANS-8.20-1991 Nuclear Criticality Safety Training

This guide specifies the material to be covered in a criticality safety course. The course is directed to:

- Workers with fissionable material,
- Supervisors of workers with fissionable material,
- Operations support personnel,
- Design personnel,
- Maintenance personnel,
- Emergency response personnel,
- Managers, and
- Administrators.

The program content shall cover:

- 1) Fission chain reactions and accident consequences,
- 2) Neutron behavior in fissioning systems,
- 3) Criticality accident history,
- 4) Control parameters,
- 5) Policy and procedures, and
- 6) Evaluation.

The order requires testing of personnel taking the course and retention of the records for at least four years.

5.2 Nuclear Regulatory Commission Regulations and Guides

Under the terms of the Atomic Energy Act of 1946 as amended, the federal government retained complete control of all materials, operations, and facilities related to the nuclear fuel cycle. The federal government set up the civilian Atomic Energy Commission to oversee the development and regulation of nuclear energy. The Atomic Energy Act of 1954 continued the control but established commercial utilization of nuclear power. The Energy Reorganization Act of 1974 split the AEC into the NRC and ERDA (Energy Research and Development Agency) which became the present DOE. The reason for the split was to separate regulation from promotion.

All federal regulations are contained in the Code of Federal Regulations. The section (called a title) devoted to nuclear energy is 10; thus, the regulations that concern us are found in 10CFR which has several parts, as shown in Table 5-1.

There are many more parts to 10CFR than shown; these listed are selected for their relevance to criticality. Parts directly applicable to the DOE begin with 200, and are extensive because they cover all other energy sources, in addition to nuclear energy.

The NRC issues regulatory guides (reg. guides) for technical guidance. These are recommendations subject to negotiation. In fact, to do other than follow them requires so much effort that the effect is compulsory for licensees; but this is not necessarily true for the DOE which is not required to conform with NRC requirements, but there has recently been a tendency to do so.

The regulatory guides are divided into 10 broad categories:

- 1) Power reactors,
- 2) Research and test reactors,
- 3) Fuels and materials facilities,
- 4) Environmental and siting,
- 5) Materials and plant protection,
- 6) Products,
- 7) Transportation,

Table 5-1 Some 10CFR Titles Applicable to the NRC	
Part	Subject
0	Conduct of employees
1	Organization and general information
2	Rules for domestic licensing proceedings
20	Standards for radiation protection
50	Licensing of production and utilization facilities
55	Operator's licenses
60	Disposal of high-level radioactive waste in geological repositories
61	Licensing requirements for land disposal of radioactive waste
70	Domestic licensing of special nuclear material (SNM)
71	Packaging of radioactive material for transport
72	Licensing requirements for the storage of spent fuel in an independent spent fuel storage location
73	Physical protection of plants and material
74	Material control and accounting of SNM
75	Safeguards on nuclear material - implementation of the US/IAEA agreement

Table 5-2 Some Division 3 Regulatory Guides Relevant to Criticality		
Number	Subject, Date	Standard
3.1	Use of borosilicate Raschig rings as neutron absorbers, 9/87	ANS-8.5
3.2	Criticality safety in operations at fuels and materials facilities, 3/86	ANS-8.1
3.9	Concrete radiation shields, 6/73	ANS-8.1
3.33	Assumptions for evaluating potential radiological consequences of criticality in a fuel reprocessing plant, 4/77	
3.34	Assumptions for evaluating potential radiological consequences of criticality in a uranium fuel fabrication plant, 7/79	
3.35	Assumptions for evaluating potential radiological consequences of criticality in a plutonium processing and fuel fabrication plant, 7/79	
3.42	Emergency planning for fuel cycle facilities and plants licensed under 10CFR parts 50 and 70, 9/79	
3.43	Criticality safety in the storage of fissile materials, 4/79	ANS-8.7
3.45	Criticality safety for steel-pipe intersections containing aqueous fissile solutions, 4/89	ANS-8.9
3.47	Criticality control and safety of homogenous plutonium-uranium fuel mixtures outside of reactors, 7/81	ANS-8.12
3.57	Administrative practices for criticality safety at fuels and materials facilities, 10/86	ANS-8.19
3.58	Criticality safety for handling, storing, and transporting LWR fuel at fuel and material facilities	ANS-8.17

- 8) Occupational health
- 9) Antitrust and financial review, and
- 10) General.

While all divisions may have some relevance, Division 3, Fuels and Materials, are the most relevant to criticality control.

Table 5-2 lists selected regulatory guides and related ANSI/ANS standards relevant to criticality control. These guides contain much of the information that has been discussed in this course and especially that discussed in the standards. This material should be referenced according to particular needs.

5.3 Department of Energy Orders

The Secretary of the DOE serves at the pleasure of the President of the United States. The DOE is responsible for nuclear, fossil, conservation renewable energy, nuclear defense programs, radioactive waste disposal, and hydroelectric power generation in the west. Fundamental research and energy research is conducted by the DOE. The central control from Washington DC is administered by eight operations offices that control national laboratories and other contractors. The rules whereby the DOE operates are called "orders", and the orders of principal concern to criticality are in some subsections of DOE Order 5480, presented in Table 5-3.

Table 5-3 Some Parts of DOE Order 5480 Relevant to Criticality	
Order Number	Subject
5480.1	Safety and health policy statement, 8/86
5480.3	Safety requirements for the packaging and transportation of hazardous materials, substance and waste, 3/87
5480.4	Environmental protection, safety, and health protection standards, 5/84
5480.5	Imposition of proposed nuclear safety requirements, 1/92
5480.6	Safety of DOE-owned nuclear reactors
5480.7	Fire protection
5480.8	Contractor occupational medical program
5480.9	Construction safety and health program
5480.11	Radiation protection for occupational workers

The order that is most relevant is DOE Order 5480.5 "Safety of Nuclear Facilities". The purpose of this order is to assure that nuclear facilities are sited, designed, constructed, operated, maintained, and decommissioned comparably to licensed nuclear facilities. Conformance with this objective means DOE and NRC requirements are consistent. They are; moreover, they are consistent with international requirements. The order defines the terms it uses and responsibilities for executing the order.

The elements of nuclear criticality safety are defined to be:

1. Process analysis - an operational readiness review shall be conducted to determine that the entire process will be subcritical under normal and reasonably expected abnormal conditions.

2. Identification of parameters preventing criticality - the basis for subcriticality shall be noted for all possible conditions. This may be a reference to established past practices. Controlling factors may be:

- a. Mass,
- b. Density,
- c. Spacing/geometry,
- d. Neutron absorbers,
- e. Moderation, and
- f. Reflection.

3. Written plans and procedures - the equipment and conduct of operations shall be in accord with written, reviewed plans and procedures.

4. Records - the operation shall provide control, signoff, and traceability of records such as plans, procedures, inspections and monitoring systems.

The process shall be such that two unlikely, independent, and concurrent changes must occur before an accidental criticality is possible. Limits for nuclear criticality shall be based on experiments. In the absence of directly applicable experiments, the limits may be calculated by a method that has been validated by experiments, provided allowance is made for uncertainties in the data and in computations.

Onsite movement of fissile materials are permitted only if such movement does not present a radiation hazard, or an accidental criticality, and after alerting fire protection, security, health physics, and other emergency personnel as appropriate.

Unirradiated fissionable material may be stored⁴ under the following conditions:

1. No non-essential combustibles in the area,
2. All operations not part of the storage operation are excluded,
3. Documented periodic inspections, test, and preventive maintenance shall be performed with a specified frequency to assure the operability of criticality prevention and safety systems, fire control, radiation protection and environmental monitoring.
4. Limits for criticality safety shall be posted conspicuously nearby locations where criticality might occur.
5. Signs shall be posted appropriately for interpretation of alarms, for evacuation routes, and for combatting fire.
6. A fire-fighting plan that is part of the site fire-fighting plan shall be prepared consistent with site emergency planning and exercised through periodic drills.
7. Auxiliary fire fighting equipment, self-contained breathing apparatus, and

⁴ These rules are not applicable to materials in process, in an assembly cell, or when the number of packages is limited in accordance with DOE Order 5480.3, radioactive waste storage or disposal facilities.

protective clothing shall be provided for protection from fire.

8. Fissionable material shall be stored in shipping containers to enhance safety.

9. All material shall be fixed to prevent geometric changes.

10. Pyrophoric material shall be rendered safe before storage.

11. All containers shall be marked to indicate the type or category of material, amount, enrichment, and radiation level.

12. The container design shall be appropriate for the type of material. Containers of material having gas buildup shall be vented in such a way that does not result in personnel exposure.

13. Materials contaminated with plutonium or U-233 shall be packaged in a closed metal container with minimum combustibles.

14. Plutonium storage facilities and containers shall be monitored periodically to assure their integrity. Procedures to detect contamination upon entering a plutonium facility shall be developed and implemented.

Summary of Chapter 5

Standards guides and orders are the codification of safe practices based on theory and experience. They are not a constraint on operations but an aid which, if followed, leads to safe operations without having to perform all of the basic investigations for potential criticality. Just follow the rules and safety is assured.

Three types of rules were discussed, the voluntary ANSI/ANS standards, the NRC law and guides which are applicable to licensees, and the DOE Orders which bear directly on DOE operations. All three are closely interconnected and have similar requirements. Basically, operations with fissile materials require identification of responsibility and criteria, process analysis extended to off-normal events, written procedures, controlled movement of fissile materials, posting of material limits, prompt reporting, investigating and correction, operation review, emergency procedures, and training.

The ANSI/ANS standards relating to criticality were reviewed, the NRC regulatory guides relevant to criticality were listed by title, and, where possible, were related to standards. The DOE requirements are contained in DOE Order 5480 - especially in order 5480.5 which was discussed by major heading.

The DOE orders provide the general rules; each facility operator must provide standard operating procedures based on the orders but adapted to the specific facility and process.

Review Questions

1. What is a Rauchig ring?
2. What are the nine administrative responsibilities delineated in standard ANS-8.1?
3. What materials become less critical when moderated?
4. If you are concerned with criticality safety in the storage of fissile materials, which regulatory guide would you consult?
5. Does DOE Order 5480.5 apply to a critical facility?
6. What six parameters are criticality controls?
7. Would you store sodium with fissile material?