# ROLPHTON NUCLEAR TRAINING CENTRE

COURSE 227

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# NUCLEAR TRAINING COURSE

# COURSE 227

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#### Nuclear Theory - Course 227

#### OBJECTIVES

At the conclusion of this course the trainee will be able to:

#### 227.00-1 Nuclear Structure

- 1. Explain and use the  $_{Z}X^{A}$  notation.
- 2. Explain the concept of binding energy.
- 3. Discuss the stability of nuclei in terms of their neutronproton ratio.
- 4. State the basic law governing radioactive decay.
- 5. State the relationship between decay constant ( $\lambda$ ) and half life ( $t\frac{1}{2}$ ).

#### 227.00-2 Neutron Reactions

- 1. Differentiate between elastic and inelastic collisions.
- 2. Explain the importance of elastic collisions to the operation of CANDU reactors.
- 3. State each of the four types of inelastic collisions, giving an example of each ( $_{Z}A^{A}$  type example is acceptable).
- 4. Differentiate between spontaneous and induced fission.
- 5. Write the equations for the formation of  $_{94}$ Pu<sup>239</sup> in our reactors.
- 6. Define:
  - a) Prompt Neutrons
  - b) Delayed Neutrons
  - c) Delayed Neutron Precursors
  - d)  $\beta$  Delayed Neutron fraction
  - e) v Neutrons Emitted per Fission
  - f) Photoneutron.
  - g) Fast neutrons
  - h) Thermal neutrons

July 1979

 Give the distribution of energy released by the fission of U-235.

# 227.00-3 <u>Neutron Cross Sections, Neutron Density and</u> Neutron Flux

- 1. Define:
  - a) Microscopic Neutron Cross Section and the units.
  - b) Macroscopic Neutron Cross Section and the units.
  - c) Neutron Density and the units.
  - d) Neutron Flux and the units.
- 2. Relate  $\sigma_a$ ,  $\sigma_f$  and  $\sigma_n$ ,  $\gamma$ .
- 3. Discuss how the microscopic cross sections of U-238 and U-235 vary with neutron energy.
- 4. Write reaction rates.
- 5. Be able to extract data from the chart of the nuclides.

### 227.00-4 Thermal Reactors

- 1. Discuss the properties of a moderator including the number of collisions required to thermalize a neutron, scattering cross section, and absorption cross section.
- 2. Define the moderating ratio.
- 3. Explain the practical significance of the fact that  $D_2O$ , compared to  $H_2O$  has a lower scattering cross section and requires more collisions to thermalize a neutron.
- 4. Discuss the effect of downgrading the moderator or heat transport fluid.
- 5. Define lattice pitch.

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- 6. Explain what "over moderated" means and why Hydro's reactor are over moderated.
- 7. Explain why increasing or decreasing the lattice pitch from its optimum value causes reactivity to change.

227.00-5 Neutron Multiplication Constant and Reactivity

- 1. Define k both in words and in terms of the six factors.
- 2. State when the word definition is not valid.
- 3. Define and explain each of the six factors in k.
- 4. Sketch a neutron life cycle using the six factors correctly.
- 5. State approximate values for each of the six factors.
- 6. Define:
  - a) Critical
  - b) Subcritical
  - c) Supercritical.
- 7. State and explain the significance of the four-factor formula for  $k_{\infty}$ .
- 8. Define and calculate values of reactivity and of reactivity worths.
- 9. Calculate values of the six factors given a neutron life cycle.

#### 227.00-6 Neutron Flux Distribution

- 1. Discuss the functions of a reflector.
- 2. Discuss the effects of a reflector.
- 3. Explain why flux flattening is desirable.
- 4. Discuss the four methods of flux flattening used.
- 5. Sketch the flux shapes showing the effect of each of the flux flattening methods.
- 6. Discuss the effect of reactor size and shape on neutron leakage.

# 227.00-7 Effect of Fuel Burnup

- 1. State and explain the units used for fuel burnup.
- 2. Explain why the combined reactivity worth due to U-235 and Pu-239 initially increases then decreases with burnup.
- 3. Explain how and why each of the four factors of  $k_\infty$  changes with fuel burnup.
- 4. Explain how and why the delayed neutron fraction ( $\beta$ ) changes with fuel burnup.

227.00-8 Changes in Reactor Power with Time

- 1. Physically explain the effect of delayed neutrons on changes in reactor power.
- 2. Given the formula,  $P(t) = \frac{\beta}{\beta \Delta k} P_0 e^{\frac{\lambda \Delta k}{\beta \Delta k}t}$ , solve calculational type problems.
- 3. Explain the concept of the prompt jump.
- 4. Define prompt criticality and explain why it is undesirable. Explain its dependence upon fuel composition and fuel burnup.

# 227.00-9 Source Neutron Effects

- State the sources of neutrons and their approximate magnitudes.
- 2. State and use the formula  $S_{\infty} = \frac{S_{0}}{1-k}$ .
- 3. Define and explain the significance of the subcritical multiplication factor.
- 4. Calculate k in a subcritical reactor given appropriate data.
- 5. State that, for a sub-critical reactor, the closer k is to one, the longer it takes for power to stabilize after a reactivity change.

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#### 227.00-10 Power and Power Measurement

- 1. Explain how thermal power is measured.
- 2. Explain why neutron power must be calibrated to thermal power.
- 3. Explain the reasons why neutron power is used for control and protection of the reactor.
- 4. State the relationship between Reactor period and rate log N. (For engineers: Prove the relationship).
- 5. Make an accurate sketch of the rundown of neutron power after a trip justifying times and power levels used.
- 6. Discuss the rundown of thermal power after shutdown.

#### 227.00-11 Fission Product Poisoning

- 1. Explain how xenon and iodine are produced in the reactor and how they are lost from the reactor.
- 2. Write the differential equations for the concentration of xenon and iodine and define each term.
- 3. State the magnitude of the production and loss terms for xenon at equilibrium in our larger reactors.
- 4. Define Xenon Load and Iodine Load.
- 5. Explain what Xenon Simulation is.
- 6. Sketch and explain the behavior of xenon after a trip from full power.
- 7. State and explain the two conditions necessary for a Xenon Oscillation.
- 8. Explain what a Xenon Oscillation is and how one may be started.
- 9. Explain why samarium growth after shutdown is not a problem.

# 227.00-12 Reactivity Effects Due to Temperature Changes

- 1. Explain why a negative fuel temperature coefficient of reactivity is desirable.
- 2. Give two undesirable effects of having a negative fuel coefficient.
- 3. Explain why the fuel temperature coefficient is more important than either the coolant or moderator temperature coefficient.
- 4. Explain why the fuel temperature coefficient is negative and why its value changes from fresh to equilibrium fuel.
- 5. Define the power coefficient and give a typical value.
- 6. Define the void coefficient.

# 227.00-13 Reactivity Control

- 1. List the various in-core reactivity worth changes, typical magnitudes of the changes, and the time period over which the changes occur.
- 2. Discuss general methods of reactivity control in terms of their effect on the six factors of k.
- Given a specific method of reactivity control (eg, Moderator Level Control) discuss its advantages and disadvantages.
- 4. List and discuss the advantages and disadvantages of each of the presently used shutdown systems.

# 227.00-14 The Approach to Critical

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- 1. Explain why the initial approach to criticality is potentially hazardous.
- 2. Explain how inverse count rate is used to predict the critical value of the controlling reactivity mechanism.

J. Crist

Nuclear Theory - Course 227

NUCLEAR STRUCTURE

# The Nucleus, Nuclear Particles

The atomic nucleus consists of Z protons and N neutrons, where Z and N are the *atomic number* and *neutron number* respectively. The total number of *nucleons* in the nucleus, that is, neutrons and protons, is equal to Z + N = A, where A is the *atomic mass number*.

A nuclear species with a given Z and a given A is called a *nuclide*. To distinguish a particular nuclide it is written in the form  $_ZX^A$  where X is the chemical symbol for the element. Nuclides with the same Z but different A are called *isotopes*. Every element has a number of isotopes - most have both stable and unstable - some have only unstable which range from 3 (hydrogen) to 26 (tin), with an average of about 10 isotopes per element.

The mass of the proton is  $1.67252 \times 10^{-27}$  kg. It carries a positive charge of  $1.60210 \times 10^{-19}$  coulombs (C), equal in magnitude to the negative charge of the electron, and it is a stable particle.

The mass of the neutron is marginally greater than that of the proton, namely  $1.67482 \times 10^{-2.7}$  kg, and it is electrically neutral. The neutron is not stable unless it is bound in a nucleus. A free neutron decays to a proton with the emission of a  $\beta^-$  particle and an antineutrino, a process which has a half-life of 12 minutes. You will see later in this course that the average lifetime of neutrons in a reactor before they are absorbed or leak from the system is no greater than a millisecond. The instability of the neutron is therefore of no consequence in reactor theory.

#### Nuclear Masses

The mass of atoms are conveniently expressed in *unified* mass units, or u. The actual mass of a nucleus is measured on the *unified* mass scale, such that the mass of the  $C^{12}$  atom is precisely 12 u, and hence 1 u = 1.660438 x  $10^{-27}$  kg.

The atomic mass of a nuclide should be distinguished from the chemical atomic weight which is the average weight of a large number of atoms of a given element. It is not quite the same as the mass of an individual atom unless the element contains a single isotope. Furthermore, you should note that the atomic weight unit on the *chemical scale* is defined as onesixteenth of the average weight of an oxygen atom in a natural mixture of stable oxygen isotopes  $(0.204\% 0^{18}, 0.037\% 0^{17}$  and the rest  $0^{16}$ ). In many calculations this slight distinction (about 3 ppm) is insignificant and the atomic mass, denoted by A, is used rather loosely.

#### Equivalence of Mass and Energy

Einstein showed that mass and energy are equivalent. The relationship between mass and energy changes may be written:

$$\Delta E = \Delta m c^2$$

where  $\Delta E$  is the energy change expressed in joules,  $\Delta m$  is the accompanying change in mass given in kilograms and c is the velocity of light, equal to 3 x 10<sup>8</sup> meters per second.

A convenient and very common unit of energy in nuclear physics is the *electron volt* (abbreviated *eV*). It is the energy gained by an electron in being accelerated through a potential difference of 1 volt.

$$1 \text{ eV} = 1.6021 \times 10^{-1.9}$$
 joule  
 $1 \text{ keV} = 10^3 \text{ eV}$   
 $1 \text{ MeV} = 10^6 \text{ eV}$ 

Using Einstein's formula it can readily be shown that converting 1 amu of mass yields  $\sim 931$  MeV of energy.

### Binding Energy

The mass of the proton is 1.00728 u, and the mass of the neutron is 1.00867 u. The actual mass of a nuclide is not equal to the total mass of its individual nucleons, the difference being called the mass defect. This mass defect is a consequence of the equivalence of mass and energy and arises from the *binding energy* of the nuclide. This is the energy required to split the nuclide into its individual component nucleons. Experimental results (Figure 1) show that except for a few light nuclides, the binding energy per nucleon in the nucleus, increases rapidly as the size of the nucleus increases up to about A = 60, but for greater values it decreases again gradually. This means that nuclei of intermediate mass are more strongly bound than the light and the heavy nuclei. Thus energy may be released by combining two light nuclei (*fusion*);

 $_{1}H^{2}$  +  $_{1}H^{3}$   $\longrightarrow$   $_{2}He^{4}$  +  $_{0}n^{1}$ 2.0147 3.0169 4.0039 1.0087 masses (u) Here 0.019 u is converted to 17.7 MeV.

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Or by splitting a heavy nucleus into two nuclei of intermediate mass (fission);

 $_{92}U^{235} + _{0}n^{1} \longrightarrow _{36}Kr^{95} + _{56}Ba^{139} + 2_{0}n^{1}$ 235.044 1.009 94.903 138.918 2.017 Mass (u)

Here 0.215 u is converted to 200.2 MeV.



#### Figure 1

# Binding Energy vs Mass Number

### Nuclear Forces

Between two electric charges of the same sign there is a repulsive force which is called a *Coulomb force*. Since nuclei may contain a large number of positive protons each repelling the other due to Coulomb forces it is clear that there must be other forces present which are attractive. These are short range *nuclear forces*. They act between all adjacent nucleons, whether n-p, n-n, or p-p, and drop off rapidly on separation of the nucleons.

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227.00-1

The lighter stable nuclei contain roughly equal numbers of neutrons and protons (eg,  ${}_{6}C^{12}$ ,  ${}_{8}O^{16}$ ,  ${}_{9}F^{12}$ ,  ${}_{11}Na^{23}$ ). As the number of protons in the nucleus increases, the long range Coulomb forces build up more rapidly than the nuclear forces which only have short range. Therefore, in order for heavier nuclei to remain intact more neutrons are required to supply binding forces between all particles to overcome the distruptive Coulomb forces. As a result, the n/p ratio required for stability gradually increases from one in light nuclei to about one and a half in heavier nuclei. This increase in the n/p ratio for stable nuclei is shown in Figure 2.

It should be noted that this is a very simple model and it cannot explain all the facts of nuclear stability or decay.



Figure 2 Neutron/Proton Ratio

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For reasons of no particular significance to us, there is a limit to the number of excess neutrons a nucleus can live with, and as a result the heavy nuclei are all unstable and there are no naturally occurring elements having a value of A greater than 238.

#### Nuclear Energy Levels

A nucleus is said to be in its ground state when the nucleons are arranged in such a way that the potential energy is a minimum. If it is not in its ground state it is said to be in an excited state and the excess of energy is called excitation energy. The potential energy does not take on a continuous range of values, but has discrete values which are termed energy levels. For heavy nuclei these energy levels have a minimum separation of about 0.1 MeV, for light nuclei this separation is much greater.

#### Radioactivity

All the naturally occurring nuclides heavier than lead (Z = 82) and a few lighter nuclides are unstable and are *naturally radioactive*. They decay by emitting either an *alpha particle* (helium nucleus) or a *beta particle* (fast electron). In most cases the resulting nucleus, or *daughter*, is produced in an excited state. It then decays to its ground state by the emission of one or more gamma photons. Usually, but not always, this occurs instantaneously, ie, within  $10^{-14}$  seconds of the formation of the daughter.

Radioactivity is governed by only one fundamental law, namely that the probability of a radionuclide decaying per unit time is constant and independent of external conditions. This constant is called the *decay constant* and is denoted by  $\lambda$ .

Thus the rate of change of single kind or radionuclide is:

$$\frac{\mathrm{d}\mathbf{N}}{\mathrm{d}\mathbf{t}} = -\lambda\mathbf{N}$$

where: N = Number Density in ATOMS/cm<sup>3</sup>  $\lambda$  = decay constant in 1/s

The solution to this simple differential equation is:

$$N(t) = N_0 e^{-\lambda t}$$

**.** .

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Thus:

The time for the number of atoms to be diminished to one half of its original value is called the half-life (t<sup>1</sup>/<sub>2</sub>).

$$N(t) = \frac{1}{2}N = N_0 e^{-\lambda (t \cdot \frac{1}{2})}$$
$$\frac{1}{2} = e^{-\lambda t \cdot \frac{1}{2}}$$
$$\ln \frac{1}{2} = -\lambda t \frac{1}{2}$$
$$t \frac{1}{2} = \frac{0.693}{\lambda}$$

The activity of a sample is simply the number of disintegrations per unit time or N. The historic unit for activity is the *Curie* (Ci), which is  $3.7 \times 10^{10}$  disintegrations per second (dps). The SI unit for activity is the *Exequenci* (Bq).

1 Bq = 1 dps

#### ASSIGNMENT

- 1. Calculate the mass defect and the binding energy for  ${}_6C^{13}$ .
- 2. In your own words, explain binding energy.
- 3. Xenon-135 has a half-life of 9.16 hours. What is its decay constant?
- 4. Sketch a graph of activity versus time, in half lives, for a radionuclide assuming that the activity is A<sub>0</sub> at time zero.

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Nuclear Theory - Course 227

NEUTRON REACTIONS

Nuclear reactions can occur as a result of collisions between various particles or gamma photons and nuclei. Charged nuclear particles, such as protons, deuterons (deuterium or  $H^2$ ) and alpha particles, need to have a large amount of energy (tens of MeV) before they are able to overcome the Coulomb repulsive forces and enter a nucleus.

Neutrons and gamma photons, however, are not charged and are therefore able to interact with nuclei very effectively, even when they have very little energy. In fact, generally speaking, there is a greater chance of a reaction occurring with low rather than high energy neutrons, because the former are in contact with the nucleus for a greater length of time.

The operation of a reactor basically depends on how neutrons react with nuclei in the reactor. It is therefore necessary to look at these reactions, called *neutron reactions*, in some detail. Although there are well over a dozen known neutron reactions, we need only consider the five that are of importance to us.

All neutron reactions can be categorized as either elastic or inelastic collisions, depending on whether kinetic energy is conserved in the collision or not.

# Elastic Collisions

Elastic collisions are those in which the total kinetic energy before the collision is equal to that after the collision.



Elastic Collision

For example, in Figure 1 a neutron with speed  $v_1$  strikes a nucleus of mass A and bounces off at lower speed  $v_2$ . The nucleus of mass A recoils with speed v, and if kinetic energy is to be conserved, the kinetic energy received by A has to be equal to that lost by the neutron. After the collision the neutron will therefore be moving at a slower speed (ie,  $v_2 < v_1$ ).

The fraction of its initial energy that the neutron loses in such a collision depends on two things:

(a) The angle at which the neutron hits,(b) The mass A of the target nucleus.

The maximum energy loss occurs when the neutron hits the nucleus head-on, and the least energy is lost in a glancing collision. The pool sharks amongst you will be well aware of this - the difference here is that the angle at which the neutron will hit the nucleus will be quite random. Consequently the angle at which it bounces off is also quite random. That is why we say that the neutron is *scattered* in the process. The term *elastic* implies the conservation of kinetic energy and therefore these collisions are described by the term *elastic scattering*.

The lighter the target nucleus is, the greater is the fraction of the energy that a neutron will lose in these collisions. Since this is the reaction with which fast neutrons are slowed down in the moderator, we want light moderator nuclei (ie, Atomic Mass Number less than 16 or so) if we are going to slow the neutrons down in as few collisions as possible. Otherwise the neutrons will travel large distances before they are slowed down thus making a physically large reactor. To emphasize this point, Table I shows the number of elastic collisions neutrons have to make in various materials to slow down from 2 MeV (the average energy with which they are produced at fission) to *thermal energy* (0.025 eV)\*.

Note that for the heavy U<sup>238</sup> nucleus, a very large number of elastic collisions would have to occur before the neutron would be slowed down to thermal energy.

\*At thermal energy, the neutrons have the same energy as the atoms or molecules with which they are colliding. At room temperature, this is about 0.025 eV.

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Number of El Fission N	astic Collisio eutrons in Var:	ons to Thermalize ious Materials
H <sup>1</sup>		18
$\mathrm{H}^{2}$	(deuterium)	25
H <sub>2</sub> O	(light water)	20
D 2 O	(heavy water)	36
C <sup>1 2</sup>	(graphite)	115
U <sup>238</sup>		2172

#### Inelastic Collisions

Instead of bouncing off, the neutron may enter a nucleus to briefly form what we call a *compound nucleus*. In such a reaction, kinetic energy is not conserved and it is therefore known as an *inelastic collision*. Basically what happens is that some of the neutron's kinetic energy is taken by the compound nucleus. As a result it becomes unstable in the sense that it cannot exist for very long in this state (ie, for no longer than about  $10^{-14}$  seconds), and the reaction that then occurs will be one of a number of alternatives described below.

1. The compound nucleus may get rid of its excess energy by emitting a neutron and a gamma photon. An example of this is shown in Figure 2. A neutron is shown entering a U-238 nucleus to form a U-239 nucleus. This immediately emits a neutron (any one) and a gamma photon to become U-238 again. The end result is still a slowing down of the neutron because the energy it has lost has been given to the gamma photon.



# Figure 2

#### Inelastic Scattering

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This reaction is known as *inelastic scattering*; "scattering" because the direction of the emitted neutron is again quite arbitrary. One of the peculiarities of this reaction is that it cannot occur unless the neutron has an initial energy of at least 0.1 MeV (this figure only applies to heavy nuclei like uranium; for lighter nuclei, around 3 MeV or more would be needed before the reaction becomes possible. These figures are based on the possible energy levels discussed in lesson 227.00-1.) From a reactor point of view, we can ignore inelastic scattering everywhere except in the fuel itself, because only there will the neutron energies be large enough for it to happen.

 An alternative to inelastic scatter is that the compound nucleus may emit either a proton or an alpha particle, and in this way form an entirely new element. Look at Figure 3, which shows such a *transmutation* of oxygen-16.





This reaction may be written as

 $n + {}_{8}O^{16} - {}_{7}N^{16} + p$ 

or you may prefer the short-hand version  $O^{1.6}(n,p)N^{1.6}$ . The N-16 is radioactive and emits high energy gamma radiation. It presents a radiation hazard in any region containing oxygen-16, that has recently been exposed to high energy neutrons. For example, oxygen-16 is present in water (either H<sub>2</sub>O or D<sub>2</sub>O), and if this water has recently flowed

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through the reactor, some of the oxygen-16 will have been changed to nitrogen-16, and this will now emit high energy gamma radiation.

Although transmutation reactions - (n,p) or (n,a) - are relatively rare, there are two more which are of interest to us:

- B<sup>10</sup>(n,α)Li<sup>7</sup>: Reactor instrumentation (ion chambers) for monitoring the neutron population in a reactor operates with this reaction. This reaction releases 2.5 MeV of energy, which shows up as kinetic energy of the helium and lithium nuclei. They lose this energy by producing a large amount of ionization in the counter, and this can easily be detected, even in the high gamma radiation background of a reactor environment. Boron is also used for reactivity control.
- He<sup>3</sup>(n,p)H<sup>3</sup>: Very sensitive reactor instrumentation makes use of this reaction, because it occurs much more readily than the one above. He-3 counters were first used in Ontario Hydro for the first start-up of the Pickering and Bruce reactors.

3. The most common neutron reaction of all is also an inelastic type of reaction. It is called *radiative* capture, because the compound nucleus has captured a neutron and it then *radiates* a gamma photon. Radiative capture can occur for practically all types of nucleus, and at all neutron energies. Generally speaking, it is more probable for slow neutrons than for fast neutrons.

An example of such a reaction is shown in Figure 4, which explains how tritium (hydrogen-3) is produced in heavy water reactors.



 $\frac{Figure 4}{Radiative Capture (n, \gamma)}$ 

Radiative capture is important for two reasons:

- (a) Non-fission neutron capture in core materials is, in a sense, undesirable. However, if the non-fission capture is with U-238 (giving U-239) there is a bonus in the subsequent transmutation of the U-239 to Pu-239. Pu-239 is a fissile nuclide and thus extends the fissile component of the fuel.
- (b) The product nucleus formed more times than not is radioactive and might present a radiation hazard. For example, corrosion products circulated by the heat transport system will be activated as they pass through the reactor core. When they later plate out in this system, the whole system becomes a radioactive hazard, and will remain so even if the reactor is shut down (ie, if the neutron source is removed). The three most troublesome activation products in our reactors are cobalt-60, manganese-56, and copper-64, and they are produced in this way.
- 4. The final reaction we are going to consider is called *fission*. The word is borrowed from the biologists, who use it to describe the breaking up of a cell into two new ones.

#### The Fission Reaction

Production of nuclear power relies on the fact that some nuclei will fission, and that energy is released during this fission process because a loss of mass occurs ( $\Delta E = \Delta mc^2$ ). There are two types of fission; *spontaneous* and *induced*.

(a) Spontaneous Fission

In this reaction, a nucleus fissions entirely spontaneously, without any external cause. It is quite a rare reaction, generally only possible for nuclei with atomic masses of around 232 amu or more. (As the atomic mass number increases, spontaneous fission becomes more and more probable. One could argue that there is an infinite number of heavy elements which do not exist, because they are not stable against spontaneous fission dacay). The table on Page 7 shows the spontaneous fission and alpha decay rates of the U-235 and U-238 isotopes.

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#### TABLE II

Spontaneous Fission And Alpha Decay Rates of Uranium

	$t_{\frac{1}{2}}(\alpha)$	$t_{\frac{1}{2}}(s.f.)$	$\alpha$ decay rate	s.f. decay rate	
	( (years)	(years)	(atoms/s/kg)	(atoms/s/kg)	
<b>U-</b> 235	7.1 x 10 <sup>8</sup>	1.2 x 10 <sup>17</sup>	79 x 10 <sup>6</sup>	0.3	
U-238	$4.5 \times 10^{9}$	5.5 x $10^{15}$	$12 \times 10^{6}$	6.9	

From this table you will be able to appreciate that spontaneous fission has no significance in the production of power. (About  $10^{-12}$ % of full power.) Nevertheless, it is important in that it represents a small source of neutrons in a reactor.

(b) Induced Fission

Certain heavy nuclei can be *induced* to fission as a result of neutron capture. In most cases the energy of the captured neutron must be very high before fission can occur, and therefore we can restrict our discussion to those nuclei which can be fissioned by neutron energies likely to be found in a reactor. In practice, we are then dealing with neutrons ranging from 10 MeV down to *thermal energies*.

#### Practical Fission Fuels

The only nuclei of practical importance to us are the U-235 and U-238 isotopes of uranium, and the Pu-239 and Pu-241 isotopes of plutonium. For all of these, except U-238, fission with thermal neutrons (*thermal fissions*) is much more probable than fission with fast neutrons (*fast fissions*). This is an important (and desirable) nuclear property, and such nuclides are said to be *fissile*. U-238, which will not fission with thermal neutrons, but which will fission with fast neutrons of energy greater than about 1.2 MeV, is merely said to be *fissionable*. It makes a small direct contribution to the power produced in a reactor, (about 3%).

Note: Fissile describes a nucleus that can be fissioned by thermal neutrons but such a nucleus can also be fissioned by neutrons of any energy.

Natural uranium only contains U-235 (0.72%) and U-238. Over a period of reactor operation, Pu-239 and also some Pu-241 will be built up in the fuel as a result of neutron capture:



Pu-239 is fissile like U-235. If it does not undergo fission, it may capture a neutron to form Pu-240. Although this is fissionable it is much more likely to capture another neutron to form fissile Pu-241. A significant fraction of the total power produced by fuel during its life in our reactors is due to fission of the fissile plutonium isotopes. We will deal with this in more detail later on in the course.

#### Fission Fragments

The fission fragments formed when spontaneous or induced fission occurs are two new nuclei. These may be any two of about 300 nuclides which are known to be formed as a result of fission.

Figure 5 (on Page 9) shows the relative frequency for nuclides of specific mass numbers produced as fission fragments. Such a curve is known as a *fission yield curve* (since two fragments are produced per fission, the area under the curve adds up to 200%). You can see that both fission fragments are likely to consist of a substantial piece of the original nucleus. They are likely to have mass numbers between 70 and 160, with those around 95 and 140 being the most probably. Note that symmetrical fission (equal fragments) is quite rare.

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Fission Yield of U-233, U-235 and Pu-239

The fission fragments are almost invariably radioactive. The reason for this is that the neutron/proton ratio of the fragments is about the same as that of the fissioned nucleus, and this is too high for stability at medium mass numbers. The fragments will therefore try to reduce their n/p ratio by successive  $\beta^-$ ,  $\gamma$  decays until stability is reached. A typical decay chain is shown in Figure 6 (on Page 10). All the members of such chains are known as *fission products*.

The half-lives of fission products range from fractions of a second to thousands of years. (It is this activity that causes so much concern in atomic bomb fall-out.) There are four important consequences of fission product production in the fuel:

(a) The fission products must be held in the fuel by encasing it in a sheath, so that they do not enter the heat transport system and hence leave the reactor core. As long as the fission products remain in the fuel and the fuel remains adequately shielded there is no biological risk.

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$$n + {}_{92}U^{235} \xrightarrow{\beta^{-}}{\gamma} {}_{39}Y^{95} \xrightarrow{\beta^{-}}{\gamma} {}_{40}Zr^{95} \xrightarrow{\beta^{-}}{\gamma} {}_{41}Nb^{95} \xrightarrow{\beta^{-}}{\gamma} {}_{42}Mo^{95}$$

# Figure 6 Fission Product Decay Chain

- (a) Continued: Since many of them have long half-lives, their presence in the heat transport system would be a radiation hazard which would prevent access to equipment even when the reactor is shut down.
- (b) Heavy shielding is required around the reactor to avoid exposure to the gamma radiation emitted by the fission products.
- (c) Fuel must be changed remotely, and special precautions must be taken in handling and storing spent fuel.
- (d) Some of the fission products have a high affinity for neutrons and thereby *poison* the reactor. The two most important poisons are Xe-135 and Sm-149. They are produced in a relatively high percentage of fissions, and they capture a significant number of neutrons.

#### Prompt and Delayed Neutron Emission

The fission fragments are produced in an excited state and will immediately emit perhaps two or three neutrons and some gamma photons. These are called *prompt neutrons* and *prompt gammas*.

Figure 7 (on Page 11) shows the energy distribution of prompt neutrons. The average energy is about 2 MeV, although the most probable energy is only 0.72 MeV.

A very small number of neutrons (less than 1%) appear long after fission occurs, and these are known as *delayed neutrons*. They arise from the radioactive decay of certain

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fission product daughters. For example:

$$\begin{array}{c} 3 \, {}_{5} \,$$

The neutron emission is instantaneous (with respect to Kr-87), but obviously occurs some time after the original fission because the Br-87 must decay first. In fact, it appears to be emitted with the 55 second half-life of Br-87.



Figure 7 Prompt Neutron Energy Spectrum

Nuclei such as Br<sup>87</sup> whose production in fission may eventually lead to the emission of a delayed neutron are known as *delayed-neutron precursors*. At the present time, it is believed that there may be as many as twenty precursors, although only about half a dozen have been positively identified. These precursors and their respective half-lives are given in Table II (on Page 12). They are usually divided into six groups according to their half-lives.

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### TABLE III

#### Delayed-Neutron Precursors

(Uncertain Quantities are Indicated by Brackets)

Precursor	Half-life (Seco	Half-life and Group (Seconds)		
Br <sup>87</sup>	54.5	Group 1		
I <sup>137</sup>	24.4	Group 2		
Br <sup>88</sup>	16.3			
I <sup>138</sup>	6.3			
Br (8 9)	4.4	Group 3		
Rb (9 3 9 4)	6			
I <sup>139</sup>	2.0			
(Cs,Sb or Te)	(1.6-2.4)	Group 4		
Br (90 92)	1.6			
Kr (9 3)	~1.5			
$(I^{1+0} + Kr?)$	0.5	Group 5		
(Br,Rb,As + ?)	0.2	Group 6		

For thermal fission of U-235, the total contribution of all the delayed neutrons (called the delayed neutron fraction;  $\beta$ ) is only 0.65% of the total neutrons produced. With Pu-239, the delayed neutron fraction is even less at 0.21%. Despite the fact that these fractions are quite small, they have a very important effect on the time dependent behaviour of thermal reactors. We shall discuss this aspect of delayed neutrons in a later lesson.

Table IV (on Page 13) gives the probability of a particular number of neutrons being emitted in the thermal fission of a U-235 nucleus. This includes both prompt and delayed neutrons.

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# TABLE IV

# Neutron Emission in Thermal Fission of U-235

Number of	Number of Cases		
Neutrons Emitted	per 1000 Fissions		
0	27		
1	158		
2	339		
3	302		
4	130		
5	34		

The <u>average</u> number of neutrons emitted per fission is a very important quantity in reactor physics. It is usually denoted by the Greek letter v ("new"). For thermal fissions of U-235, v = 2.43. (Fast fissions, ie, fissions caused by fast neutrons, usually produce marginally more neutrons.) It is also interesting to compare the number of neutrons released per thermal fission of Pu-239 and Pu-241 since both of these plutonium isotopes build up in our fuel after a while.

#### TABLE V

Values of v for Thermal Fissions

Fissile Nucleus	ν
U -235	2.43
Pu-239	2.89
Pu-241	2.93

Energy Release From Fission

About 200 MeV of energy is liberated when a nucleus fissions. The exact value slightly depends on the fissile nucleus and on the fission fragments produced. The energy can be calculated as follows:

Consider the example given in Figure 6 on page 10:

 $_{92}U^{235} + n \longrightarrow_{38}Sr^{95} + _{54}Xe^{139} + 2n$ 

Total mass before fission = 235.044 + 1.009= 236.053 amuTotal mass after fission = 94.903 + 138.918 + 2.018 = 235.839 amu

Loss in mass = 0.214 amu

This corresponds to almost 200 MeV. A summary of how this energy is distributed is given in Table VI.

#### TABLE VI

#### Approximate Distribution of Fission Energy Release in U-235

Kinetic energy of lighter fission fragment	100	MeV
Kinetic energy of heavier fission fragment	69	MeV
Energy of prompt neutrons	5	MeV
Energy of prompt y rays	6	MeV
$\beta$ particle energy gradually released from fission products	7	MeV
$\gamma$ ray energy gradually released from fission products	6	MeV
Neutrinos (energy escapes from reactor)		MeV
Total	204	MeV

This is not a complete account of all the energy released in the reactor. Some of the neutrons even after losing all their kinetic energy may produce  $(n,\gamma)$  reactions with materials in the reactor, and up to about 8 MeV may be released in such reactions. The total amount of energy produced in a reactor per fission may therefore depend to a slight extent on the form of the reactor, but it is always within a few MeV of 200 MeV.

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Not all of this 200 MeV of energy from fission is useful or desirable. The principal useful heat is due to the kinetic energy of the fission fragments. This shows up as heating of the fuel from which the heat is transferred to the heat transport fluid. Most of the neutron and about one third of the gamma energy (~5-6% of the total) shows up as heating of the moderator. This is essentially wase heat which must be rejected. The energy due to decay of the fission products makes up about 7% of the total fission energy. This has a major effect on reactor design since this energy shows up as heat for a long time after essentially all fissioning has ceased. Because of this decay heat we must have a shutdown cooling system for normal shutdown conditions and an emergency core cooling system in the event that normal cooling is lost. As demonstrated at 3 Mile Island, even when shut down a reactor is still producing about 1% of its full thermal power.

#### Reactor Power and Fuel Consumption

The 200 MeV released in one fission is not of much practical value because it is minute. In fact, 1 watt of power requires  $3.1 \times 10^{10}$  fission every second.

One Megawatt steady power requires 3.1 X  $10^{16}$  fissions every second continuusly. 3.1 X  $10^{16}$  atoms of U-235 have a mass of:

 $\frac{3.1 \times 10^{16} \times 235}{6.023 \times 10^{25}} = 1.21 \times 10^{-8} \text{ kg}$ 

Therefore, to produce 1 Megawatt-day of energy from fission requires the complete fissioning of:

 $1.21 \times 10^{-8} \pm 24 \times 3600 = 1.0 \times 10^{-3} \text{ kg} = 1.0 \text{ g} \text{ U-}235$ 

The first requirement for producing useful power from the fission process is that enough U-235 nuclei must be available for fissioning. This requirement is met by installing sufficient U-235 in the reactor in the form of fuel rods. If natural uranium is used, of which 0.72% is U-235, then about 140 g of uranium would be used to produce 1 Megawatt-day of energy. This assumes that all the U-235 could be fissioned. In practice this is not so, because some U-235 ( $\sim$ 14%) is consumed in (n, $\gamma$ ) reactions. As a result, 165 g of natural uranium would be used.

For example, a Pickering reactor at full power generates 1744 MW from fission (540 MW gross electrical power). It would therefore use about 290 kg of natural uranium a day on this basis. Because Pu-239 (and Pu241) is produced in the fuel after a while, this contributes substantially to energy production, and the amount of fuel used is consequently smaller.

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#### Production of Photoneutrons

Prompt and delayed neutrons are produced as a result of fission. If no further fissions occur, no more prompt or delayed neutrons will be produced. This is not the case with photoneutrons.

Photoneutrons are peculiar to reactors with heavy water moderator or heat transport fluids. They are produced when photons with energies greater than 2.2 MeV are captured by deuterium nuclei:

 $\gamma + _{1}H^{2} \longrightarrow _{1}H^{1} + n$ 

After the reactor has been operating for a while, it will have built up in the fuel an inventory of fission products whose gamma decay photons have an energy greater than 2.2 MeV. Even when the reactor is shut down, this photoneutron source will persist because the gamma rays from decaying fission products can still produce photoneutrons in any heavy water present in the core. Even if the moderator has been dumped, heavy water will always be in the core as heat transport fluid. Therefore in our heavy water cooled reactors we always have a relatively large neutron source (compared to the spontaneous fission source) with which to start the reactor up again after a shutdown.

#### ASSIGNMENT

- 1. Explain why we use materials with a low atomic mass for moderators.
- Table II shows the spontaneous fission rate for U-238 as
   6.9 fissions/s/kg. Is this fission rate of any signifigance? Explain your answer.
- 3. How long will it take delayed neutrons to come into equilibrium after a power change?

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#### Nuclear Theory - Course 227

NEUTRON CROSS SECTIONS, NEUTRON DENSITY AND NEUTRON FLUX

Neutron Cross Sections

Let us have a look at the various reactions a neutron can undergo with a U-235 nucleus:

As mentioned in lesson 227.00-2:

- 1. If the neutron energy is greater than 0.1 MeV, inelastic scattering may occur. If the neutron energy is less than this, there is no chance of this reaction happening.
- 2. The neutron may just bounce off (elastic scattering), and this can happen at all neutron energies.
- 3. The neutron may be captured (radiative capture).
- 4. The neutron may cause fission.

Radiative capture and fission are much more likely for slow neutrons than for fast neutrons, and fission is always more probable than radiative capture.

Thus we are always comparing the chances in favour of the various reactions taking place. It is the probability of a certain reaction occurring that is important. Some reactions are more probable with some nuclei than with others or more probable with some neutron energies than with others. Because these reactions are concerned with a neutron striking a target, namely a nucleus, the probability that a certain reaction will occur is measured in terms of an area, called the *Neutron Cross-Section*.

To understand this cross-section better, imagine the neutrons as being bullets shot at the target in Figure 1, instead of at a nucleus. When the neutron misses the target altogether, no reaction takes place. The areas of the various rings on the target represent the chance of various reactions occurring. Thus the area, d, of the complete disc, being the easiest to hit, represents the probability of the easiest reaction occurring. The area to the outside of the single-hatched ring, c, represents the probability of the next easiest reaction occurring. Area b represents the probability of the third easiest reaction occurring and area a, of the bull's eye, the probability of the most difficult reaction occurring, since the bull's eye is the most difficult to hit. The areas of these rings can be such that the probability of an area being hit by a bullet is equal

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to the probability of a reaction occurring between the neutron and the nucleus. The area of the ring is, then, the crosssection for that particular reaction. Because these crosssections apply specifically to individual nuclei, they are known as *microscopic* cross-sections.



# Figure 1

Needless to say the ring areas are extremely small, being of the order of  $10^{-24}$ ,  $10^{-23}$  or  $10^{-22}$  square centimeters. A special unit, called the *barn*, is therefore used to describe these cross-sections.

$$1 \text{ barn} = 10^{-24} \text{ cm}^2$$

The barn is of the same sort of size as the physical target area  $(\pi r^2)$  presented by a medium sized nucleus.

If a reaction has a large cross section, say 100 b, it will occur much more frequently than one that has a small cross-section, say 0.1 b. In fact, it is exactly 1000 times as likely.

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As was pointed out earlier, when there are a number of possible reactions with a given nucleus, each one would have its own cross-section. The Greek letter  $\sigma$  (sigma) is used as the symbol for the microscopic cross-section, and so:

 $\sigma_{f}$  = fission cross section  $\sigma_{a}$  = absorption cross section  $\sigma_{n,\gamma}$  = radiative capture cross section  $\sigma_{i}$  = inelastic scattering cross section

 $\sigma_{s}$  = elastic scattering cross section

 $\sigma_a$  is usually the radiative capture cross section, ie,  $\sigma_n, \gamma$ . Only in those few cases where fission is also possible, (ie,  $\sigma_f \neq 0$ ),  $\sigma_a$  would include  $\sigma_f$  and  $\sigma_n, \gamma$  since a neutron is absorbed in both cases;

ie,  $\sigma_a = \sigma_f + \sigma_n, \gamma$ ,

since both fission and radiative capture involve a complete absorption and loss of the neutron. So, to repeat, for nuclides with  $\sigma_f = 0$ ,  $\sigma_a$  is merely  $\sigma_n$ , $\gamma$ .

Cross sections depend very much on the neutron energy. Generally speaking, they are a lot larger at low energies than at high energies. For example, the fission cross-section  $\sigma_f$  for U-235 for neutrons of thermal energy is 580 b, whereas it is only just over 1 b at MeV. In other words, fission of U-235 is about 500 times as likely for thermal neutrons than for fast neutrons. This very nicely illustrates what the moderator does for us.

For your interest, Table I lists the thermal neutron crosssections of fuel nuclei. It might be quite instructive to have a look at these numbers and see what we can make of them.

In the table  $\sigma_a$  is shown as  $\sigma_f + \sigma_n, \gamma$ , since both processes involve a complete absorption of the neutron.

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(taken	from	Atomic	Energy	Review	(IAEA), 1969,	Vol 7, 1	No 4, p.3)
			σf	σ <sub>n</sub> ,γ	σ <sub>a</sub>	് <sub>5</sub>	V
	U-233	3 53	30.6	47.0	577.6	10.7	2.487
	U-235	5 58	30.2	98.3	678.5	17.6	2.430
	U-238	}	0	2.71	2.71	$\sim$ 10	0
	Nat.	U	4.18	3.40	7.58	$\sim$ 10	
	Pu-23	39 74	41.6	271.3	1012.9	8.5	2.890
	Pu-24	1 100	07.3	368.1	1375.4	12.0	2.934

#### TABLE I

Thermal Neutron Cross Sections of Fuel Atoms (in Barns) taken from Atomic Energy Review (IAEA), 1969, Vol 7, No 4, p.3)

Only 86% of the thermal neutrons absorbed by U-235 cause fission. You can see that this is just the fraction  $\sigma_f/\sigma_a$ . Note also that U-233 gives the greatest percentage of fission per neutron absorbed ( $\sigma_f/\sigma_a = 92$ %); this is a very desirable aspect of U-233, and for this reason it may well be used in future reactors.

The values for natural uranium were obtained by using 99.3% of the U-238 values and 0.7% of the U-235 values. Looking at the table, you can see that for natural uranium  $\sigma_f = 4.18$  b,  $\sigma_n, \gamma = 3.40$  b and hence  $\sigma_a = 7.58$  b. This means that for every, say, 758 thermal neutrons absorbed in natural uranium, 418 will cause fission. Since these fissions can only occur in U-235, we will get v = 2.43 new neutrons produced per fission. The 418 fissions will therefore generate 418 x 2.43 = 1016 new neutrons. This means that for every thermal neutron absorbed in natural uranium fuel, we will on average get back 1016/758 = 1.34 new ones.

In our reactors this is a sufficient number because we have relatively few neutron losses in reactor materials (in other words, the absorption cross-sections of the reactor materials we use are small enough). However, the U.S. reactors use a light water moderator. Light water has an absorption cross-section that is almost 700 times greater than that of heavy water. As a result, the light water absorbs so many neutrons that 1.34 new neutrons for every neutron absorbed in the fuel are not enough. They therefore use *enriched* fuel, ie, the U-235 concentration is greater than the naturally occuring one of 0.72%. You might

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like to work out for yourself (using the values given in Table I) what difference an enrichment of 2% U-235 makes (2% enrichment means 2% U-235 and 98% U-238).

Now that we have described what cross sections are, let us take this discussion a little further. Imagine 1 cm<sup>3</sup> cube of a certain kind of material, and let this cube contain n thermal neutrons. These n neutrons are all zipping around inside the



cube with velocity v and they will make collisions with the nuclei sitting there. We will assume that there are N nuclei in the 1 cm<sup>3</sup> cube, and that their absorption ( $n,\gamma$ ) cross-section is  $\sigma_a$ . It turns out that the number of neutrons interacting (ie, being absorbed) per second is given by:

$$R = nv.N\sigma_{e}$$
(1)

R is called the *reaction rate*. Intuitively you can see that the expression for R seems reasonable, because:

- (a) The larger n, the more neutrons will make collisions
- (b) The larger their velocity, the more nuclei they will get to hit in a certain time,
- (c) The larger the number of nuclei present (N)
  the more will be hit, and
- (d) The larger the cross-section, the greater is the probability of getting a hit.

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This result is quite general, and if we were to use  $\sigma_{\rm f}$  instead of  $\sigma_{\rm a}$  in the expression, R would be the number of fissions per second. The quantities N and  $\sigma$  are both characteristic of the so-called target material, and therefore they are often combined to form the

macroscopic cross section 
$$\Sigma = N\sigma$$
 (2)

 $\Sigma$  is the capital  $\sigma$ , and note the spelling macroscopic instead of miroscopic.

The units of  $\Sigma$  will be cm<sup>-1</sup>. For example, let us work out  $\Sigma_a$  for natural uranium. N is 0.048 x  $10^{24}$  nuclei/cm<sup>3</sup> and from Table 1  $\sigma_a$  is seen to be 7.58 barns.

$$\therefore \Sigma_a = N\sigma$$
  
= 0.048 x 10<sup>24</sup> ( $\frac{1}{cm^3}$ ) x 7.58 x 10<sup>-24</sup> (cm<sup>2</sup>)  
= 0.36 cm<sup>-1</sup>

What does this mean? Well, please take my word for it that  $1/\Sigma_a$ , which is a distance, is the average distance a neutron will travel before being absorbed in the material. That is, thermal neutrons zipping around in natural uranium will travel an average distance of 1/0.36 - 2.8 cm before they are absorbed.

Appendix B gives the values of  $\Sigma_a$  for all of the elements and for light and heavy water. The cross-sections apply to thermal neutrons only. This table has been included for interest's sake only, but it does bring out which materials have high neutron capture cross-sections and which don't.

To return now to equation (1), we can write it as

$$R = nv.\Sigma$$
(3)

n is the number of neutrons per  $cm^3$ . We call this the *neutron* density, for rather obvious reasons.

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nv is called the *neutron flux*. It is the total distance travelled by all the n neutrons in  $1 \text{ cm}^3$  in one second, since each of them will cover a distance v. The Greek letter  $\phi$  (phi) is always used for neutron flux. Its units are

It represents the total neutron tracklength per unit volume per unit time. We therefore end up with

$$\mathbf{R} = \phi \Sigma$$

To see what sort of use these ideas have, let us look at an operating reactor that has an average thermal neutron density of 100 million, ie,  $n = 10^8 \text{ cm}^{-3}$ . This is a typical figure. The speed of thermal neutrons is still quite high, it is in fact 2.2 km/s, that is 2.2 x  $10^5 \text{ cm} \cdot \text{s}^{-1}$  (or 5000 m.p.h., if you like to look at it that way). Therefore, this reactor has an average neutron flux

$$nv = \phi = 2.2 \times 10^{13} n.cm^{-2}s^{-1}$$

If the reactor uses natural uranium, the the absorption rate per  $\rm cm^3$  of fuel is

$$\phi \Sigma_a = 2.2 \times 10^{13} \times 0.36 = 7 \times 10^{12} \text{ s}^{-1}$$

If the reactor contains  $10^6$  cm<sup>3</sup> of fuel (ie,  $\perp$  m<sup>3</sup>) then there will be

 $7 \times 10^{12} \times 10^{6} = 7 \times 10^{18}$  neutron captures per second.

Going back to Table I, you can see that 4.18 in every 7.58 neutrons captured will cause fission. We will then have

 $\frac{4.18}{7.58} \times 7 \times 10^{18} = 3.8 \times 10^{18}$  fissions per second.

We saw earlier that  $3.1 \times 10^{10}$  fissions per second will produce 1 watt, therefore in this case the reactor is producing

 $\frac{3.8 \times 10^{18}}{3.1 \times 10^{10}}$  watts = 123 x 10<sup>6</sup> W = <u>123 MW</u> (thermal)

## Chart of the Nuclides

We have now covered all the material necessary to use the chart of the Nuclides which is included as Appendix C. For those self-studying this course, a few minutes spent studying the explanation of the chart will be time well spent.

## Variation in Cross Sections

As mentioned, neutron cross sections are highly energy dependant. The variation in cross section is not a simple function of neutron energy. Figure 2 shows the variation of the absorption cross section of U-238 with energy. Of particular interest here are the pronounced peaks between  $\sim 5$  eV and  $\sim 1$  keV. These are called resonance absorption peaks and the corresponding energies resonance energy. The cross sections are so high in these regions that a large portion of the neutrons at these energies will be absorbed.



# Figure 2

All cross sections have some energy dependance. At low energies most cross sections are inversely proportional to the neutron velocity, ie,

$$\sigma \propto \frac{1}{v} \text{ or } \frac{1}{\sqrt{\mathbf{z}}}$$

Variations from this normal behavior will be covered when they have an effect on overall behavior of the reactor.



### Neutron energy - eV

Variation of the absorption cross section of U-235 with neutron energy.

# Figure 3

### ASSIGNMENT

- 1. Explain what a microscopic neutron cross section is.
- 2. If 100 thermal neutrons were absorbed by natural uranium, how many fast neutrons would be produced? What is the significance of your answer?
- 3. Using the Chart of the Nuclides, trace the radioactive decay of U-238 to a stable nuclide.
- 4. If a thermal neutron interacts with a U-235 nucleus, calculate the probability that the interaction will be a scattering reaction.

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Nuclear Theory - Course 227

THERMAL REACTORS (BASIC DESIGN)

When a U-235 nucleus fissions an average of 2.5 neutrons are released in addition to the energy. This suggests that these neutrons could be used to cause additional fissions thus creating a chain reaction. Assume we have natural uranium (0.72% U-235) as fuel. Let us start with some numbers of fission neutrons (say 1000). These neutrons have an average energy of about 2 MeV (see Table 4, lesson 227.00-2). Since the fission cross-section of U-235 is about 1 barn at this energy it seems reasonable to "slow" the neutrons to low energy where the fission cross-section is much higher (580 barns for a neutron energy of 0.025 eV). We can rapidly reduce the energy of a neutron by having it undergo elastic collisions with a light nucleus (H, He, C, etc). As a result we obtain neutrons which are in thermal equilibrium with their surroundings and are therefore called "thermal neutrons". At a temperature of 20°C a thermal neutron has an energy of 0.025 eV.

During the slowing down process the neutrons pass through the resonance absorption energies of the U-238. If the fuel and moderator were intimately mixed (homogeneously) too many of the neutrons would suffer resonance capture and a chain reaction could not be sustained. If, however, we separate the fuel into discrete locations within the moderator, ie, using fuel channels, the neutrons can slow down away from the U-238 thus avoiding resonance capture.

At thermal energies the neutrons diffuse around until they are absorbed by the fuel, or leak out of the reactor, or are absorbed by something other than the fuel (moderator, fuel sheath, pressure tubes, etc). By careful choice of reactor materials we can limit the non-fuel or parasitic absorption. In addition we can reduce leakage by careful design of the size and shape of the reactor. Of the neutrons absorbed by the fuel, some will cause fission while others will simply undergo radiative capture. If enough of the neutrons cause fissions to give us the 1000 neutrons we started out with then we have a selfsustained chain reaction. We can define a neutron multiplication factor (k) for this reaction as:

 $k = \frac{\text{number of neutrons in one generation}}{\text{number of neutrons in the preceding generation}} *$ 

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<sup>\*</sup>Note that this definition is only valid when the effects of source neutrons (photoneutrons and spontaneous fission neutrons) are negligible.

For the chain reaction to be self-sustaining  $k \ge 1$ . We will deal with k in more detail in the next lesson. The remainder of this lesson will deal with moderator properties, reducing neutron leakage, and spacing of the fuel channels.

#### Moderator Properties

The primary objective of a moderator is a lot easier to express than to achieve: the fission neutrons must be slowed down to thermal energies without being absorbed. Let us examine the latter aspect first:

There are two possibilities:- the neutrons can be absorbed by the moderator atoms themselves or by fuel atoms, and this can occur anywhere in the energy range from  $\sim 2$  MeV (fission neutrons) down to 0.025 eV (thermal neutrons). Absorption by moderator atoms can obviously be minimized by choosing a moderator with a sufficiently low absorption cross-section, but for fuel the argument is rather more subtle.

Recall that U-238 exhibits a number of severe absorption peaks between 5 eV and 1 keV. It is essential to minimize resonance capture, and one way of doing this is to ensure that, in the slowing down process, the neutron energy loss per collision is as high as possible. For example, consider the moderators in Figure 1 (for the sake of simplicity the resonances have been smoothed out).





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Moderator 2 thermalizes the neutrons in far fewer collisions that Moderator 1. This means that the neutrons in Moderator 2 will spend less time in the resonance energy region, and will therefore also have less chance of colliding with U-238 while they have this energy. If they do collide, they will almost certainly be captured. The conclusion is that there will be less resonance capture in U-238 with Moderator 2 than with Moderator 1.

#### Slowing Down Mechanism

Having established that we want to slow the neutrons down in as few collisions as possible we shall now examine how this might be achieved.

There are two slowing down mechanisms:

- (1) inelastic scattering (with fuel nuclei)
- (2) elastic scattering (with moderator nuclei)

(Inelastic scattering with moderator nuclei is not possible because the neutron energies are too low, and even with uranium nuclei it is only possible down to about 100 keV. In any case, it is relatively unimportant. Elastic scattering with fuel nuclei may be ignored, because the energy loss per collision is negligible).

In an elastic collision the energy lost by a neutron depends on the mass of the target nucleus and the angle of collision. Since the angle of collision is totally random, a mathematical function can account for its effect. By manipulating equations for conservation of momentum and conservation of kinetic energy we could prove that, the most energy is lost when a neutron collides with a target of equal mass and that for targets of general mass A the energy lost is a simple function of the mass of the target.

It takes a number of collisions for a fast (2 MeV) neutron to slow to thermal energy (0.025 eV) and the larger the mass of the target nucleus, the larger the number of collisions required. This is due to the fact that a smaller portion of the neutron's energy is lost per collision. The mathematical function used to express this is the mean logarithmic energy decrement  $\xi$  (xi).

$$N\xi = Ln \quad \frac{E_i}{E_f}$$

Where:

and

N = number of collisions to thermalize  $E_i$  = initial neutron energy  $E_f$  = final neutron energy N = total E loss going from  $E_i$  to  $E_f$ 

 $\xi$  = mean log energy decrement

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Table I shows the accurate values of  $\xi$  of a number of light materials which might be suitable as moderators.

## TABLE I

# Mean Logarithmic Decrements

	Ę	Collisions to Thermalize
H <sup>1</sup> *	1.000	18
H <sup>2</sup> *	0.725	25
He <sup>4</sup> *	0.425	43
Be <sup>9</sup>	0.206	83
C <sup>1 2</sup>	0.158	115
H <sub>2</sub> O	0.927	20
D <sub>2</sub> O	0.510	36
BeO 0.174		105
		i

\*Gases at STP

### Slowing Down Power and Moderating Ratios

A small number of collisions to thermalize is obviously desirable, but this is of no use on its own unless the collisions actually occur. This implies that the probability of a collision must be high, that is  $\Sigma_s$  should be large. Recall that:

$$\Sigma_s = \sigma_s N$$

This immediately rules out gases as moderators, because N would be too small for the neutrons to be slowed down within a reasonable distance.

The overall effectiveness of a material for slowing down neutrons is measured by the product  $\xi \Sigma_s$  which is known as the Slowing Down Power.

Table II shows the slowing down powers of the solid and liquid moderators. The value of the slowing down power is also shown to demonstrate the unsuitability of a gas.

## TABLE II

Slowing	Down	Powers	and	Moderating	Ratios
2					

	ξ	$\Sigma_{s}(cm^{-1})^{(a)}$	ξ <sup>Σ</sup> s	Σa	$\xi \Sigma_s / \Sigma_a$
He <sup>(b)</sup>	0.425	21x10 <sup>-6</sup>	9x10 <sup>-6</sup>	? very small	? large
Ве	0.206	0.74	0.15	1.17x10 <sup>-3</sup>	130
C <sup>(c)</sup>	0.158	0.38	0.06	0.38x10 <sup>-3</sup>	160
BeO	0.174	0.69	0.12	0.68x10-3	180
H <sub>2</sub> O	0.927	1.47	1.36	22x10- <sup>3</sup>	60
D <sub>2</sub> O	0.510	0.35	0.18	0.33x10 <sup>-4(d)</sup>	5500 <sup>(d)</sup>
D <sub>2</sub> O	0.510	0.35	0.18	0.88x10 <sup>-4</sup> (e)	2047 <sup>(e)</sup>
D <sub>2</sub> O	0.510	0.35	0.18	2.53x10 <sup>-4 (f)</sup>	712 <sup>(f)</sup>

(a)  $\Sigma_s$  values of epithermal neutrons

- (ie, between  $\sim$ l and  $\sim$ 1000 eV)
- (b) at S.T.P.
- (c) reactor-grade graphite
- (d) 100% pure  $D_2O$
- (e) 99.75% D<sub>2</sub>O
- (f) 99.0% D<sub>2</sub>O

Not only must the moderator be effective in slowing down neutrons, but it must also have a small capture cross-section. Neutrons are slowed down to decrease radiative captures compared to fission captures, and obviously the whole purpose of moderation would be defeated if the moderator nuclei themselves captured neutrons.

A reasonable indication of the overall quality of a moderator is the *Moderating Ratio*, which combines the slowing down power and the macroscopic capture cross section:

Moderating Ratio = 
$$\frac{\xi \Sigma_s}{\Sigma_a}$$

We are now in a position to draw some interesting conclusions from Table I.

 $H_2O$  has excellent slowing down properties, and is often used as a fast neutron shield (neutrons must be slowed down before they can be absorbed. Why?) Unfortunately its  $\Sigma_a$  is too high to permit its use as a moderator for natural uranium fuel, and enrichment is necessary.

Be, BeO and graphite have lower values of  $\Sigma_a$ , and can be used with natural uranium fuel provided the fuel is in metal form. The use of natural uranium compounds with more attractive physical and chemical properties (such as UO<sub>2</sub> or UC) is not feasible with these moderators, because of the reduction in the concentration of uranium atoms. The British line of power reactors used a graphite moderator with natural uranium metal fuel (their earlier Magnox stations), and in the early '60s they changed to graphite with enriched UO<sub>2</sub> fuel (the AGR stations).

In the U.S., an abundance of U-235 produced for weapons and a tradition of using it in nuclear submarines led to all out development of light water reactors with relatively highly enriched fuel and a relatively poor moderator.

You can see from Table I that heavy water is by far the best moderator as far as its nuclear properties go, and of course its use was adopted for the CANDU line of reactors (CANDU = CANadian-Deuterium-Uranium). Its  $\Sigma_a$  is so low that natural uranium can even be used in compound form as  $UO_2$ .

The substance used as a moderator must be very pure. It is usually used, in a reactor, in larger amounts than any other material, eg, the volume of carbon in a graphite moderated reactor is 70 to 80 times that of the fuel. A very small amount of impurity in a moderator can substantially increase its capture cross-section. The addition of 1 boron atom to every million graphite atoms would increase the capture crosssection of graphite by 25%

For the same reason the isotopic purity of  $D_2O$  must be kept high. The addition of 0.25% H<sub>2</sub>O to pure  $D_2O$  more than doubles the capture cross-section. Thus, the isotopic purity of moderator  $D_2O$  is kept at 99.75% by weight or better. This is known as *reactor-grade*  $D_2O$ . As you might surmize from the moderating ratio, it would be difficult if not impossible to keep the reactor critical if the isotopic were allowed to drop to 99%. For a more practical approach, Figure 2 shows the change in reactivity with moderator isotopic. Downgrading of the moderator by <u>only</u> 0.1% will introduce about -4 mk. (By contrast downgrading of the heat transport fluid is less important simply because the volume is much less. Thus, downgrading of the heat transport fluid to 95% isotopic will introduce about -5 mk.)



## Figure 2

# Effect of Moderator or P.H.T. Downgrading

# The Diffusion of Neutrons Through the Moderator

Many parameters which determine the design of a reactor are dependent on the way neutrons are slowed down and diffuse in the moderator.

Neutrons diffuse through a material as a result of being scattered by nuclei. Neutrons virtually never collide with each other because the neutron density is so much smaller than the atomic density. The treatment of neutron diffusion, which is a process similar to the diffusion of electrons in a metal, is too complicated to include in this course, and we shall therefore restrict ourselves to the pictorial representation in Figure 3.



#### Figure 3

#### Neutron Diffusion In A Moderator

A fission neutron born at A is thermalized after several collisions and arrives at B. The average distance between A and B in a Candu reactor is about 25 cm. After slowing down the neutron diffuses to C where it is absorbed. The distance BC is about 30 cm. These are known as the mean "crow-flight" distances and are straight line displacements not total distance travelled.

The crow-flight slowing down distance determines the optimum distance between adjacent fuel channels. This spacing is called the *Lattice Pitch*. Figure 4 shows the approximate variation of k (the neutron multiplication factor), with lattice pitch.

Note that if the lattice pitch varies in either direction from its optimum value, k will decrease. If the pitch is increased we have extra moderator and some neutrons are being unnecessarily absorbed by the moderator before they reach the fuel. In this case the reactor is said to be overmoderated. If the pitch is decreased, we don't have enough moderator and some neutrons reach the fuel while still at resonance energy thus more are absorbed wastefully by the U-238 resonances. In this case the reactor is undermoderated.

All of Ontario Hydro's reactors are overmoderated. The reasons for this are physical rather than nuclear. The pressure tubes must have sufficient separation to allow the



fueling machine access to the end fitting on either end of the fuel channel and the calandria tubes must have sufficient separation to allow space horizontally and vertically for control mechanism guide tubes.

Overmoderating our reactors by a small amount has very little effect as you can see by looking at the decrease in k on Figure 4 for the PGSA reactors.

The behaviour of k with Lattice Pitch has an additional benefit in that any significant accidental rearrangement of the reactor structure makes the reactor less reactive, for example, core disassembly in a melt down.

#### ASSIGNMENT

- 1. What is the practical significance of the fact that  $D_2O$  is poorer at slowing down neutrons than  $H_2O$ ?
- 2. Explain why changing the lattice pitch from the optimum value causes a decrease in reactivity. Why is this a safety feature?

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## Nuclear Theory - Course 227

NEUTRON MULTIPLICATION FACTOR AND REACTIVITY

In the previous lesson the neutron multiplication factor (k) was defined as:

 $k = \frac{\text{no. of neutrons in one generation}}{\text{no. of neutrons in the preceeding generation}}$ 

This definition is only valid if the neutron power is high enough that the effect of source neutrons (photoneutrons and spontaneous fission neutrons) may be ignored and if k itself is not changing. A more precise way to define k is as the product of six factors, each of which represents a possible fate for a member of the neutron population. Thus:

$$\mathbf{k} = \epsilon p \eta f \Lambda f \Lambda t$$

Where:

- $\epsilon$  (epsilon) = Fast Fission Factor. The factor by which the fast neutrons population increases due to fast fission.
  - $\varepsilon = \frac{\text{No. of neutrons from}}{\text{No. of neutrons from}} + \frac{\text{No. of neutrons from}}{\text{fast fission}}$
- A typical value is about 1.03 for natural uranium fuel

p = Resonance Escape Probability. The probability
 that a neutron will not undergo resonance
 capture in U-238 while slowing down.

No. of neutrons leaving resonance energy range p = No. of neutrons entering

the resonance energy range

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A typical value is about 0.9 for natural uranium fuel.

η (eta) = Reproduction Factor. The number of neutrons produced by thermal fission per neutron absorbed by the fuel.

$$\eta = v \frac{\sum_{f=1}^{fuel}}{\sum_{a}^{fuel}} = v \frac{\sum_{f=1}^{fuel}}{\int_{f=1}^{fuel}}$$

- A typical value is about 1.2 for natural uranium fuel.
  - f = Thermal Utilization. The fraction of the thermal neutrons absorbed by the fuel compared to neutrons absorbed in the whole reactor.

$$= \frac{\sum_{a}^{\Sigma} fuel}{\sum_{a}^{\Sigma} total reactor}$$

A typical value is about 0.95 for a CANDU reactor core. Note: Fuel must be defined the same way for both  $\eta \& f$ .

- A<sub>f</sub> = Fast Non-leakage Probability. The probability
  that a fast neutron won't leak out of the
  reactor. A typical value is about 0.995.
- $\Lambda_t$  = Thermal Non-leakage probability. The probability that a thermal neutron won't leak out of the reactor. A typical value is about 0.98.

The first four factors, which depend only on the materials of construction, are frequently grouped together and called the multiplication factor for an infinite reactor  $(k_m)$ .

 $\mathbf{k}_{\infty} = \epsilon \mathrm{pnf}$ 

f

This is normally referred to as the "four-factor formula".

The last two factors are leakage factors which depend on the size and shape (ie, the geometry) of the reactor. Figure 1 shows how each of the factor relates to the neutron life cycle.



Neutron Life Cycle

# Figure 1

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A thermal neutron which is absorbed by the fuel may be absorbed by fissile material (U-235 or Pu-239) or by nonfissile material (Fission Products, U-238, etc). If it is absorbed by fissile material it may undergo radiative capture or cause fission. If it causes fission,  $\nu$  fast neutrons will be produced. The reproduction factor ( $\eta$ ) accounts for all of this. Thus for N thermal neutrons absorbed by the fuel N $\eta$  fast neutrons are produced.

As U-238, Pu-239, and U-235 all have small but finite fission cross-sections for fast neutrons, the fast neutrons can cause additional fissioning to take place. This results in an increase ( $\approx 3$ %) in the fast neutron population. The fast fission factor ( $\epsilon$ ) accounts for this increase in the fast neutron population. Thus for Nn fast neutrons from thermal fission we get  $\epsilon$ Nn fast neutrons from fast and thermal fission.

The two factors  $\varepsilon$  and  $\eta$  are essentially properties of the fuel and the magnitude of the product  $\varepsilon\eta$  fixes the tolerable limits of the other factors which can be regarded as design variables. That is the product  $pf\Lambda_f^{\Lambda}t \stackrel{>}{=} \frac{1}{\varepsilon\eta}$ .

While slowing down the fast neutrons may reach the boundary of the reactor and leak out. To account for this reduction in the population we have the fast non-leakage probability  $(\Lambda_f)$ .

The fast neutrons may also suffer resonance capture while slowing through the resonance energy range. The resonance escape probability (p) accounts for this. Thus for Nen fast neutrons starting the slowing down process NenpAf neutrons reach thermal energy.

A certain percentage of the thermal neutron population will diffuse to the boundary and leak out. We use the thermal non-leakage probability  $(\Lambda_{th})$  to account for this loss.

The remaining thermal neutrons will either be absorbed by the fuel or by the core material. The thermal utilization factor (f) accounts for this. Thus for  $NepnfA_f^A$  thermal neutrons,  $NepnfA_f^A$  are absorbed by the fuel.

From Figure 1 you can see that if we divide the number of neutrons in the (i + 1)<sup>th</sup> generation by the number in the i<sup>th</sup> generation we have:

 $k = \frac{N \epsilon p \eta f \Lambda f \Lambda t}{N} = \epsilon p \eta f \Lambda f \Lambda t$ 

When k = 1 the reactor is said to be critical. If k is unity and the effects of source neutrons are negligible, neutron power will be constant in a critical reactor. It is important to realize that a reactor may be critical at any power level and that telling someone that a reactor is critical tells them nothing about the reactors power output. By analogy; if I tell you that a car is not accelerating, do you know how fast it is going? If we want to increase power we must make k greater than one by reducing the losses, with respect to fission, of neutrons. The reactor is then said to be supercritical. Power will continue to increase as long as k is maintained at a value greater than one.

To reduce power we must increase the losses of neutrons thus making k less than one. The reactor then is said to be subcritical and power will decrease until the source neutrons become significant. (This point will be covered in detail in lesson 227.00-9.)

### Reactivity

A reactor is critical when k = 1. The factor that determines how subcritical or supercritical a reactor may be, is the amount by which k differs from 1.

A quantity called reactivity, is used to describe changes in k which are called reactivity changes. Reactivity is defined as:

$$\frac{k-1}{k}$$

For values of k close to 1 (eg, 0.98 to 1.02) which easily encompasses our normal operating range.

Reactivity may be approximated as

 $\Delta \mathbf{k} = \mathbf{k} - \mathbf{1}$ 

This is the accepted meaning of reactivity in Hydro.

The reactivity changes that are made for normal reactor control are always quite small, and they are measured in a unit called the milli-k or mk. (This is not strictly a unit but is a fraction, 1 mk is the same as 0.1%, ie, 0.001).

For Example: k = 1.002  $\Delta k = k - 1$  = 1.002 - 1= 0.002 or 2 mk

A typical CANDU reactivity control system such as the liquid control zone at Bruce and Pickering have a range of about 6 mk.

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## ASSIGNMENT

- 1. Put your text and your notes away. Now, write the six factor formula, define each of the terms, and sketch the neutron life cycle with the terms used correctly.
- 2. Calculate the exact value of reactivity for k = 0.95.
- 3. Calculate each of the six factors for the neutron life cycle shown below.



Nuclear Theory - Course 227

NEUTRON FLUX DISTRIBUTION

From neutron diffusion theory it is possible to derive the steady state flux distribution in a reactor. Since the flux is not normally the same everywhere in a reactor, its distribution or shape is obviously of importance because it will determine the distribution of power generated in the core. Generally the flux has a maximum at the centre of the core, and drops off to zero outside the moderator volume because there is no thermal neutron source there.

In a cylindrical reactor, shown below, there are two directions along which the flux distribution is considered. These are the axial direction,  $\phi_z$ , and the radial direction,  $\phi_r$ , from the centre of the reactor.



Figure 1



The thermal neutron flux  $\phi_{(r,z)}$  at a point (r,z) in the cylinder is given by:

 $\Phi_{(r,z)} = \phi_m J_o \left(\frac{2.405r}{R}\right) \cos \left(\frac{\pi z}{H}\right)$ 

where  $\phi_m$  is the maximum flux. It occurs at the point 0. J<sub>o</sub> (2.405 r/R) gives the radial flux distribution. It is a special function, namely a zero order Bessel function. Fortunately it is only marginally different from a cosine function.

Unfortunately the ratio of the average flux  $(\phi_{avg})$  to the maximum flux  $(\phi_{max})$  is only 27.5%. The total power output of the reactor depends on  $\phi_{avg}$ .

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One way of increasing the average flux, hence the power, is to increase the maximum flux,  $\phi_{max}$ . However,  $\phi_{max}$  is normally limited by the maximum fuel heat rating, and this will be reached first at the centre of the reactor. One way in which the rest of the fuel can be made to contribute "its share", is to deliberately flatten the flux distribution over part of the reactor. For example, if the average flux can be increased from 27.5% to 55% of the maximum, the same reactor can supply twice the power.

The justification for flux flattening is therefore an economic one. We will discuss flux flattening later in this lesson but first we need to look at the loss of neutrons due to leakage from the reactor.

#### Neutron Leakage

Knowing that Candu fuel is used in a reactor, let me raise the question "Can a single fuel bundle be made critical in a vat of heavy water?" The answer is no, because too many of the fission neutrons escape from the fuel never to return (ie, the non-leakage probabilities  $\Lambda_f$  and  $\Lambda_{th}$  are too low). Now let us assemble more and more fuel bundles, properly spaced, until the reactor is critical. The minimum size of this assembly of fuel and moderator which will yield a selfsustaining chain reaction is called the critical size. For fixed reactor materials and spacing, the critical size is determined by:

- 1) the shape of the reactor
- 2) what happens to a neutron at the reactor boundary.

To illustrate the importance of shape, assume that eighteen fuel bundles assembled as shown below, with a  $D_2O$  moderator and optimum lattice pitch (25.5 cm), make a critical mass.



Figure 2

- 2 -

Now ask yourself "Would the same eighteen fuel bundles be critical in a single tube surrounded by a  $D_2O$  moderator?"



Figure 3

The answer is again no, because the leakage is far too great.

Both the effects of size and shape can be combined by observing that the smaller the surface area of the core per unit volume of the core, the smaller will be the leakage. Based on this observation you would build a large spherical reactor (see 327.00-1).

The astute mechanical designers amongst you will recognize that a spherical reactor would be very difficult (ie, expensive) to construct, therefore, we use the next best shape - a cylinder in which the height is approximately equal to the diameter. The size of the reactor is essentially determined by how large a turbine-generator unit the station is going to have.

All our reactors except NPD are quite large and thus have minimal leakage (Pickering and Bruce,  $D \approx H \approx 6$  m: NPD,  $D \approx H \approx 3.5$  m). Neutron leakage can be further reduced by surrounding the core with a substance which scatters or reflects neutrons back into the core. Such a substance is known as a *reflector*. An additional benefit of using a reflector is that it produces a flatter flux distribution, and therefore better fuel utilization.

#### The Function of the Reflector

Figure 4 on the next page shows the function of a reflector diagrammatically. Figure 4(a) shows a "bare" core with many neutrons escaping. In Figure 4(b) a substance has been placed around the core to reflect most of the neutrons back into the core.



#### Figure 4

### The Function of the Reflector

It is evident that, with the reflector, more neutrons are available for fission because the leakage is smaller. Therefore, the core size does not have to be increased as much in order for the reactor to go critical. That is, the critical size of a reflected core is smaller than that of a bare core. Alternatively, if the size of the core is kept the same, higher fuel burnups can be achieved with consequent reduction in fuel costs.

## Reflector Properties

Neutrons are reflected back into the core as a result of scatterings with reflector nuclei; hence, a material with a high scattering cross-section is desirable. It is equally desirable that the reflector not absorb too many neutrons (low absorption cross-section). These are the same things that we desire from a moderator.

For this reason, the reflector usually is just an extension of the moderator (approximately 70 cm for our large reactors). This has the advantage of (a) simplifying the design of the reactor vessel and (b) obviating the need for a separate reflector system.

## The Effects of Adding a Reflector

The effects of placing a reflector around the core can be summarized as follows:

1. The thermal flux is "flattened" radially, ie, the ratio of average flux to maximum flux is increased. This is illustrated in Figure 5. The hump in the curve is due to the fact that fast neutrons escape into the reflector and are thermalized there. They are not as likely to be absorbed there as they are in the core.



Figure 5

The Effect of a Reflector on the Thermal Flux Distribution

- 2. Because of the higher flux at the edge of the core, there is much better utilization of fuel in the outer regions. This fuel, in the outer regions of the core, now contributes much more to the total power production.
- 3. The neutrons reflected back into the core are now available for fission. This means that the minimum critical size of the reactor is reduced. Alternatively, if the core size is maintained, the reflector makes additional reactivity available for fuel burnup.

## Flux Flattening

For maximum power output from a given reactor, it is desirable that each fuel bundle contribute equally to the total power output. As we have shown, in an unreflected (bare) reactor the average flux ( $\phi_{avg}$ ) is only 27.5% of the maximum flux ( $\phi_{max}$ ). Thus the average fuel bundle is producing only one

quarter of the power it could safely produce (assuming the bundle which is exposed to the peak flux is producing the maximum power it can safely produce).

To improve this situation we attempt to flatten the flux, ie, reduce the peak to average flux ratio:

$$\left(\frac{\phi_{avg}}{\phi_{max}}\right)$$
.

For our reactors four methods of flux flattening are used:

- 1) Reflector (previously discussed)
- 2) Bi-directional refuelling
- 3) Adjuster rods
- 4) Differential burnup.

## Bi-directional Refuelling

If adjacent fuel channels are fuelled in opposite directions, as they are in our reactors, an automatic flux flattening arises in the axial direction. The effect is illustrated in Figure 6.



### Figure 6

Effect of Bi-Directional Refueling

The effect is due to the fact that the newer fuel (at the input end of the channel) will generate a higher flux than the highly burned up fuel at the exit end. How much flattening is obtained in this way actually depends on how many bundles are replaced during refueling. From the point of flux flattening,

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the fewer the better; however, other considerations (discussed in Reactor, Boilers & Auxiliaries, 133.60-2) largely determine the number of bundles replaced. Even with the present refueling schemes (8 or 10 out of 12 bundles at Pickering and 8 or 10 out of 13 at Bruce) some flux flattening is obtained. Additionally, bi-directional refueling prevents the development of the undesirable flux distribution which would result from unidirectional, partial channel, refueling (shown in Figure 7).





#### Adjuster Rods

Adjusters are rods of a neutron absorbing material which are inserted into the central regions of the reactor to suppress the flux peak which normally occurs there. The name adjusters comes from their function (ie, adjusting flux) and they should not be confused with control absorbers. Adjusters affect both the radial and axial flux. Figure 8 shows the radial flux distribution in a reactor with adjusters and one without. Note that both flux curves are drawn with the same maximum flux; thus, the reactor with adjusters gives a higher power output for the same maximum flux.

The Pickering-A reactors use 18 adjuster rods (shown in Figure 9) constructed of Cobalt. When Cobalt absorbs a neutron it becomes Co-60  $({}_{27}\text{Co}^{59} + {}_{0}\text{n}^1 \rightarrow {}_{27}\text{Co}^{60} + \gamma)$ . The adjusters are replaced periodically and the Co-60 is processed and marketed by AECL. The designs of Bruce-B, Pickering-B, and Darlington include the use of 21 stainless steel adjuster rods.

Inasmuch as adjuster rods are normally inserted in the reactor at full power, they represent a negative reactivity contribution. To overcome this we must reduce the fuel burnup by approximately 10%. This is reflected in slightly higher fuel costs.

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## Figure 8

Effect of Adjuster Rods

In addition to flattening the flux, adjuster rods are withdrawn to add positive reactivity for Xenon override.

## Differential Burnup

Differential burnup is a method of flux flattening used at Douglas Point and Bruce-A which avoids incurring the fuel burnup loss experienced due to adjusters. For this purpose the reactor is divided into two regions radially as shown in Figure 9.

The fuel in Zone I is allowed to burnout approximately 1.5 times as much as the fuel in Zone II. With more highly burned out fuel in the centre of the core there is less fissioning taking place, hence lower flux. The effect is shown in Figure 10. Note that differential fueling gives flux flattening only in the radial direction.

Table I lists the present Ontario Hydro Reactors, the methods of flux flattening used and the resultant peak to average flux ratios.







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	Reflector	Bi-Directional Fuelling	Adjusters	Differential Burnup	¢avg ¢max
NPD	axial & radial	$\checkmark$			42%
Douglas Point	radial	$\checkmark$		$\checkmark$	50%
Pickering-A	radial	$\checkmark$	$\checkmark$		57%
Pickering-B	radial	$\checkmark$	$\checkmark$		60%
Bruce-A	radial	$\checkmark$		$\checkmark$	∿59%
Bruce-B	radial	$\checkmark$	$\checkmark$		∿60%
Darlington	radial	$\checkmark$	$\checkmark$	$\checkmark$	∿60%

TABLE I

The expression:

$$P = \frac{\overline{\phi} \cdot M}{3 \times 10^{12}}$$

relates the total power output P (in MW thermal) to the total mass of uranium fuel M (in Mg U) for an average thermal flux  $\overline{\phi}$ . You will appreciate that increasing  $\overline{\phi}$  without increasing the maximum flux  $\phi_m$  has enormous economic benefits. For instance, the first four Pickering units cost 765 million dollars. Without any flux flattening at all,  $\overline{\phi}/\phi_m$  would have been around 27%, ie, for roughly the same investment\* we would have got less than half the installed capacity.

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<sup>\*</sup>You wouldn't have had to pay for the  $D_2O$  reflector and the adjuster rods, and any loss in fuel burnup not off-set by cobalt-60 production.

# ASSIGNMENT

- 1. Using sketches explain why a reflector is more important at NPD than it is at Pickering.
- 2. Why do we flatten the flux in Ontario Hydro's reactors?
- 3. Explain how each of the methods of flux flattening works.

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Nuclear Theory - Course 227

EFFECT OF FUEL BURNUP

The changes in the composition of the fuel as it is depleted give rise to a number of effects which may be described under the following headings:

- 1) Long Term Reactivity Effects
- 2) Reactor Kinetics Effects
- 3) Neutron Flux Distribution Effects

Burnup Units

Before discussing the effects of fuel burnup we must first look at the commonly used units. Burnup is expressed either in terms of:

- (a) the total heat energy extracted per unit weight of fuel, preferably expressed in MWh/kgU. (Note: MWh is thermal energy not electrical energy.)
- (b) the total neutron exposure (flux x time), of the fuel, normally expressed in neutrons/kilobarn (n/kb). This is a convenient but illogically named unit arrived at by multiplying flux (<u>neutron·cm</u>) cm<sup>3</sup>·s

by time (s) and getting units of neutrons/ $cm^2$ . Therefore:

 $l n/kb = \frac{1 \text{ neutron}}{10^3 \times 10^{-24} \text{ cm}^2} = 10^{21} \frac{\text{neutrons}}{10^2 \text{ cm}^2}$ 

This is more properly expressed as  $10^{21} \frac{\text{neutron cm}}{\text{cm}^3}$ ,

the total neutron track length per unit volume.

#### Long Term Reactivity Effects

The composition of the fuel will change quite significantly during its life in the reactor. There are two predominant effects: the burnup of fissile U-235 and the conversion of non-fissile U-238 into fissile plutonium.

The rate at which these occur depends on the neutron flux, because the rate  $\frac{dN}{dt}$  of neutron capture by nuclides

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per unit volume, is given by

$$\frac{dN}{dt} = N\sigma_a \phi$$

Where N is the number of nuclides/unit volume.  $\sigma_a$  is the absorption cross-section per nuclide.  $\phi$  is the neutron flux.

For U-235 exposed to a typical flux of  $10^{14}$   $\frac{\text{neutron} \cdot \text{cm}}{\text{cm}^3 \cdot \text{s}}$ 

it takes about four months to burnout half of the U-235 initially present. Fortunately the burnout of U-235 is offset by the conversion of U-238 to fissile Pu-239 by the following scheme:

 $92U^{238} + 0n^1 \rightarrow 92U^{239} + \gamma$ 

 $92U^{239}$   $\frac{1}{2} = 24 \text{ min}^{239} \gamma + \beta + 93Np^{239}$ 

 $_{93}Np^{239} \frac{1}{t_2} = 56 h^{>} \gamma + \beta + _{94}Pu^{239}$ 

The Pu-239 that is produced will eventually build up to equilibrium when its rate of production will be equal to its rate of removal ( $\sigma_f = 742b$ ,  $\sigma_n, \gamma = 271b$ ). This will be at an irradiation of about 3 n/kb. The Pu-240 formed by neutron capture has properties very similar to U-238, but if it captures another neutron it will form fissile Pu-241 ( $\sigma_f = 1007b$ ,  $\sigma_n\gamma = 368b$ ).

Therefore after a long period of reactor operation, power will be produced from fission of U-235, Pu-239 and Pu-241.

Figure 1 shows the concentration of these nuclides as a function of total flux exposure. Table I presents the same data using both burnup units. Note that flux exposure and energy extracted are not linearly related due to the variation in the fission cross section of the fuel.

Of equal significance to overall long term reactivity is the buildup of Pu-240 and neutron absorbing fission products (other than Xenon which will be considered separately). Figure 2 shows the approximate reactivity variation due to the major factors just discussed.

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CONCENTRATION (g/kg U)

# TABLE 1

Burnup	Data*
--------	-------

n/kb	MWh/kgU	U-235 (g/kgU)	Pu-239 (g/kgU)	Pu-241 (g/kgU)
0	0	7.20	0	0
0.2	19	6.37	0.60	0.002
0.4	39	5.62	1.10	0.009
0.6	59	4.90	1.48	0.025
0.8	79	4.30	1.77	0.049
1.0	100	3.76	1.98	0.078
1.2	120	3.32	2.14	0.107
1.4	140	2.90	2.25	0.145
1.6	159	2.56	2.33	0.177
1.8	179	2.26	2.39	0.211
2.0	198	1.98	2.43	0.245
2.2	216	1.74	2.46	0.278
2.4	235	1.54	2.48	0.309
2.6	253	1.35	2.49	0.338
2.8	271	1.18	2.50	0.366
3.0	289	1.03	2.50	0.393

\* The values shown in this table strictly speaking apply only to the Pickering reactors, but they will be correct to within a percent or so for all natural uranium,  $D_2O$  moderated reactors.



Initially the burnup of U-235 and its replacement by a smaller number of Pu-239 nuclei ( 8 Pu-239 atoms created for each 10 U-235 atoms burned up) leads to an increase in reactivity. This is due to the higher fission cross section of Pu-239. ( $\sigma_f$  (Pu-239) = 742b;  $\sigma_f$  (U-235) = 580b).

At higher irradiations the U-235 is still being removed, but the buildup of Pu-239 becomes less rapid as it approaches its equilibrium level when the production of Pu-239 will equal the removal due to absorption. Consequently at high irradiations the reduction in the number of fissile nuclei causes a reduction in reactivity.

The build up of Pu-240 produces a large negative reactivity contribution due to significant neutron absorption ( $\sigma_a = 280$  b). This is partially offset by the buildup of fissile Pu-241. There is an initial rapid decrease of fission product reactivity due mainly to Sm-149 ( $\sigma_a = 41,800$  b) which reaches an equilibrium after about 300 hours of operation. This rapid decrease is followed by a nearly linear decrease due to the continuing creation of mildly neutron absorbing fission products.

It is also useful to examine how the four factors of the infinite multiplication factor  $k_{\infty}$  vary with burnup.

Figure 3 is a graph of the predicted variation of  $k_{\infty}$  and the four factors taken from the Bruce Design Manual. First note that neither the fast fission factor ( $\varepsilon$ ) nor the resonance escape probability (p) show any significant variation and can be assumed to be constant with respect to fuel burnup. This is due to the fact that most fast fission and resonance capture takes place in U-238 which constitutes ~99% of the fuel whether it is fresh or equilibrium fuel.


The most important variation is in the reproduction factor ( $\eta$ ). Recall from lesson 227.00-5 that:

$$\eta = v \quad \frac{\sum_{f} f}{fuel}$$

$$\sum_{a} fuel$$

While all of the parameters in n change with irradiation, the most important variations are :

1. Initial decrease due to Sm-149

- 2. Increase (to about 0.5 n/kb) due to the buildup of Pu-239
- Continuing decrease after 0.5 n/kb -due to burnout of U-235 and the buildup of Pu-240 and fission products.

The thermal utilization (f) increases slightly due to increasing absorption in the fuel relative to the core structural materials. (Note that the buildup of Pu-239, Pu-240, Pu-241, and fissions products all lead to increased absorption by the fuel.)

Clearly at some point in time the value of k will go below one and we no longer have a useful reactor. Normally we target our reactors to operate at full power with small amounts of positive reactivity (typically  $\approx 5$  mk) available in addition to the Xenon override capability. Figure 4 is a plot of the actual excess reactivity at Pickering unit #1 for the initial fuel charge.

After ~180 full power days daily onpower refueling was started to maintain the target reactivity. At this point in time the reactor is said to be at an equilibrium fuel condition. From this point onward refuelling takes place on a daily basis at a rate equal to the burnout rate; somewhere between 8 and 18 bundles per day. Prior to this the reactor is said to be in the fresh fuel condition. (Note: When speaking of the entire reactor we refer to fresh or equilibrium fuel, when refering to an individual fuel bundle it is either fresh or irradiated.

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227.00-7

FIGURE 4

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#### Reactor Kinetics Effects

The main effect on reactor kinetics is the change in the delayed neutron fraction ( $\beta$ ) with fuel burnup. Recall from lesson 227.00-2 that  $\beta$ (U-235) = 0.65% and  $\beta$ (Pu-239) = 0.21%. The importance of this change will become apparent in the lesson on Reactor Kinetics.

## Neutron Flux Distribution Effects

As discussed in the previous lesson both bidirectional refuelling and differential fuelling are useful for flux flattening due to the different characteristics of fresh and irradiated fuel.

## Fuel Management Calculations

The Fuel Engineer on the station is responsible for ensuring that as far as possible, the optimum fuel cycle is used.

In other words, that maximum reactor power be maintained with minimum fuel cost.

Various computer programs exist which are capable of following the histories of the bundles in the core. For example, such programs calculate the expected axial and radial power distributions, the burnup of each bundle in the core and the excess reactivity available. The validity of these calculations can be checked by comparing the power distributions put out by the program with those obtained from the flow rates and temperature increases ( $\Delta$ T) in the various channels. If there are large discrepancies, the physics data of the program is modified by intelligent guesses until eventually the agreement between theory and practice is close enough.

The Fuel Engineer uses the output of such a program (typically this might be run monthly) to help him decide which channels to fuel when. Since the core is usually divided into a number of annular zones of roughly equal ratings, (eg, there are 8 such zones at Pickering), the fuelling rates per zone can easily be derived. Even so, no rigid fuelling pattern is used; the following criteria would have to be considered.

- Discharge of highest burnup fuel (this information is obtained from the program).
- (2) High reactivity gain per channel fuelling (mainly intuitive).

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- (3) No fuelling in high temperature areas if derating is likely to be necessary (the reactor control computer will print out a temperature matrix).
- (4) Symmetry
- (5) Equal numbers fuelled per reactor quadrant (Douglas Point) or per liquid control zone (Pickering & Bruce).
- (6) Alternate East and West fuelling.
- (7) Effect on neighbouring channels.
- (8) Experimental bundles.
- (9) Priority must be given to channels known to contain failed fuel.

After a channel has been fuelled, the corresponding changes in bundle positions will have to be input for the next run of the computer program. If the axial flux distribution in the reactor is fairly flat, it might well be expedient to fuel in so-called 8 or 10 bundle shifts. Figure 5 shows the changes in bundle positions for an 8 bundle shift at Pickering.





FIGURE 5

At Pickering for example, the adequacy of the fuelling program is assessed with the following guidelines:

- (1) Reactivity variations within normal range liquid zone level control.
- (2) Maximum channel outlet temperature well below first temperature alarm, and a minimum number of channels above a specified power level.
- (3) Minimum flux tilt, ie, zone levels similar.
- (4) Channel burnup evenly distributed within each annular zone no significant over-irradiation of fuel.

We have control over items 2, 3 and 4 but for item 1 we are at the mercy of fuelling machine performance.

### ASSIGNMENT

- Explain (using appropriate formulas) the formation of Pu-239 & Pu-241 in a CANDU reactor.
- 2. Explain how and why the reproduction factor (n) changes from fresh to equilibrium fuel in a CANDU reactor.
- 3. Could the state of the fuel (ie, fresh or equilibrium) make any difference in the ability to override Xenon? Explain your answer.
- 4. Using Figure 1 calculate the total fissile content of the fuel at exit from a Pickering reactor as a percentage of the initial fissile content. Inasmuch as the percentage you have calculated is rather high, explain why the fuel isn't left in the reactor longer.

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227.00-8

Nuclear Theory - Course 227

CHANGES IN REACTOR POWER WITH TIME

Reactor kinetics is the study of how neutron power changes with time. As a preface to this discussion it must be recognized that neutron density (n), neutron flux ( $\phi$ ), and neutron power (P) are all related by physical or design constants such that they all behave in a similar manner. Their relationships are:

 $P = EV\Sigma_{f}\phi$  $\phi = nv$ 

where:

Ρ	=	neutron power
Ε	=	energy released per fission
Σf	=	the macroscopic fission cross section
φ	=	neutron flux
V	=	volume of the reactor
n	Ξ	neutron density
v	=	average neutron velocity

Prompt Kinetics

First we shall examine the behaviour of a reactor without delayed neutrons. The change of the neutron density in one generation is:

 $\Delta n = kn - n$ 

where:

kn = the neutrons in one generation

n = the neutrons in the preceding generation

k = neutron multiplication factor

The time period over which this takes place ( $\Delta t$ ) is one neutron lifetime (1).

Thus:

$$\frac{\Delta n}{\Delta t} = \frac{kn - n}{\ell}$$

or:

$$\frac{dn}{dt} = \frac{kn - n}{\ell} = \frac{n}{\ell} (k - 1) \times \frac{k}{k}$$

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$$\frac{dn}{dt} = \frac{nk}{\ell} \frac{k-1}{k}$$

Recall that:

$$\frac{k-1}{k} = \Delta k$$

Normally k is very close to one so that:

 $\frac{\ell}{k} \approx \ell$ 

We can now rewrite the equation as:

 $\frac{\mathrm{d}n}{\mathrm{d}t} = \frac{n}{\ell} \Delta k$ 

The solution to this equation is:

$$n(t) = n_0 e^{\frac{\Delta kt}{\ell}}$$

where:

n = original neutron density

Since both neutron flux and neutron power behave in a similar manner we can write:

$$P(t) = P_{o}e^{\frac{\Delta kt}{\lambda}}$$
(1)

Equation (1) shows that power changes exponentially with time and that the rate of change of power depends on the reactivity ( $\Delta k$ ) and the neutron lifetime ( $\ell$ ).

## Reactor Period

In operating reactors it is convenient to have an indication of how long it takes for power to change by a given amount (e.g. how long it takes for power to double or increase by a certain percentage). The most common measure in Candu reactor is how long it takes power to increase by a factor of e\*. This time interval is called the reactor period T (tau).

\* e is the base for natural logarithms and is used simply for mathematical convenience. e = 2.7183

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To illustrate what the reactor period is, consider the reactor power after one reactor period (ie, t = T).

 $P(T) = eP_0$  (ie, power has increased by a factor of e)

Also:

$$P(T) = P_{O}e^{\frac{\Delta k}{\ell}}$$

Therefore:

Clearly:

$$1 = \frac{\Delta k}{\ell} T$$

and:

 $T = \frac{\ell}{\Delta k}$  (reactor period for a reactor with only prompt neutrons)

Thus we can rewrite equation (1) as:

$$P(t) = P_0 e^{\frac{t}{T}}$$
(2)

Equation (2) is a valid expression for power as a function of time considering that we have only prompt neutrons.

To gain a feel for what this means, consider such a reactor with  $\Delta k = 0.5 \text{ mk}$ ;

$$T = \frac{0.001 \text{ s}}{0.0005} = 2 \text{ s}$$

This means that, with the reactor only slightly supercritical (k = 1.0005), power is increasing by a factor of e ( $\sim$  270%) every 2 seconds. That is about 176% per second\*.

\*Do not confuse this with rate log N which would be 50%/s for this example.

This is an unacceptable rate of power change because it would be mechanically impossible to build a regulating system which could respond to such changes rapidly enough to safely control the reactor. Fortunately the fission process produces delayed neutrons which radically alter the time response from that of prompt neutrons alone.

## Effect of Delayed Neutrons

In Level 3 Nuclear Theory we simply assumed that the delayed neutrons increased the average neutron lifetime. This simple treatment is not only calculationally inaccurate but it also fails to predict the physical way in which delayed neutrons affect the reactor. A more complex treatment is required for deeper understanding.

Again we will look at the time rate of change of the neutron density  $\frac{dn}{dt}$ , which can be written as:

$$\frac{dn}{dt} = \frac{kn}{\ell} \begin{pmatrix} 1 - \beta \end{pmatrix} + \frac{\lambda C}{Term 2} - \frac{n}{\ell}$$
Term 1 Term 2 Term 3

where:

- Term 1 represents the production of prompt neutron in the present generation
- Term 2 represents the production of delayed neutrons in the present generation
- Term 3 represents the total neutrons in the preceding generation
  - $\lambda$  = delayed neutron precursor decay constant
  - C = delayed neutron precursor concentration

With some mathematical manipulation:

$$\frac{dn}{dt} = \frac{n}{\ell} \left( k (1 - \beta) - 1 \right) + \lambda C$$
$$= \frac{kn}{\ell} \left( \frac{k - k \beta - 1}{k} \right) + \lambda C$$
$$= \frac{kn}{\ell} \left( \frac{k - 1}{k} - \beta \right) + \lambda C$$

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Finally:

$$\frac{dn}{dt} = \frac{kn}{\ell} (\Delta k - \beta) + \lambda C$$
(3)

Equation (3) partially describes how 'n' is changing; however,  $\lambda C$  is not a constant. We continually create delayed neutron precursors while other precursors, created earlier, decay to give us delayed neutrons. So we must describe how the precursor concentration changes:

$$\frac{dC}{dt} = \frac{kn}{\lambda} \beta - \lambda C$$

precursor precursor creation decay rate rate

Recalling that there are six groups of precursors you can see that we would have seven simultaneous differential equations to solve.

For a calculationally accurate prediction of power changes, these equations are solved on a computer. For the purpose of understanding; however, we assume an average behaviour of the delayed neutrons which reduces the problem to solving two equations with a solution of the form:

$$P(t) = A_0 e^{a_0 t} + A_1 e^{a_1 t}$$

If we assume:

- a)  $\Delta k < \beta$
- b) a step change in  $\Delta k$  occurs at time zero
- neutron density was constant prior to the insertion of reactivity.

The solution (with certain approximations) is:

$$P(t) = P_0 \begin{pmatrix} \frac{\lambda \Delta k}{\beta - \Delta k} t & -\frac{\beta - \Delta k}{2} t \\ \frac{\beta}{\beta - \Delta k} e & -\frac{\Delta k}{\beta - \Delta k} e \end{pmatrix}$$
(4)  
Ist Term 2nd Term

A mathematical and graphical solution for a typical set of conditions is shown on the next page.



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From this example you can see that the second term of equation (4) dies away rapidly and can usually be ignored.

If we do neglect the second term we have:

$$P(t) = \frac{\beta}{\beta - \Delta k} P_{o}e$$
(5)

Graphically this simplified equation is:



This behaviour is called a "prompt jump" followed by a "stable period" where the stable period (T) =  $\frac{\beta - \Delta k}{\lambda \Delta k}$ .

# Physical Effect of Delayed Neutrons

To understand what is physically happening we will look at a simple numerical example using a greatly exaggerated value for  $\beta$ . This is done only for numerical simplicity and in no way alters the qualitative results.

Assume:

$$\beta = .1$$
  
 $\Delta k = .05$   
 $n_0 = 1000$ 

Prior to the reactivity insertion:



The chain reaction is being maintained at a level of 1000 neutrons per generation. Now insert 50 mk of reactivity such that k = 1.05:



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Even though we create 105 precursors we get only 100 delayed neutrons from the precursor bank since it contains precursors produced earlier.

This chain proceeds as follows:



If we assume the output of the precursor bank does not change for a second, we have time for one thousand prompt generations in which time the series will converge to:



Thus in a very short time period we get a jump in the prompt neutron level but power can not increase beyond a certain point until more precursors start to decay. Therefore, after the prompt jump, the rate of power increase is determined by the decay rate of the delayed neutron precursors. Calculating the magnitude of the prompt jump for this problem using equation (5).

$$P = \frac{\beta}{\beta - \Delta k} P_0 = \frac{.1}{.1 - .05} 1000 = 2000$$

## Approximate Numerical Effect of Delayed Neutrons

Going back to the problem we solved without delayed neutrons we shall now see what the effect of delayed neutrons really is.

Consider a reactor with U-235 as the fuel then:

 $\beta = 0.0065$  $\lambda \approx 0.1 \text{ s}^{-1}$ 

The reactor period with  $\Delta k = 0.5$  mk is:

$$T = \frac{\beta - \Delta k}{\lambda \Delta k} = \frac{0.0065 - 0.0005}{(0.1) \ (0.0005)}$$

= 120 s

and the overall power function is:

$$P(t) = P_0 \frac{\beta}{\beta - \Delta k} e^{\frac{t}{T}}$$
$$\frac{P(t)}{P_0} = 1.08 e^{\frac{t}{120}}$$

After the initial prompt jump to 108% of  $P_{\rm O}$  power increases with a period of 120 s.

Figure 2 shows the power rise for a step insertion of reactivity. As you can see the average lifetime approximation (from Level 3) fails to predict the rapid initial rise in power caused by the multiplication of prompt neutrons. This rapid rise in power is an important consideration in the design of all reactivity mechanisms. In order to limit any rapid increase in power all reactivity mechanisms are designed to limit the rate of reactivity addition.

## Prompt Criticality

It may have occurred to you to ask why we restricted  $\Delta \mathbf{k}$  to being less than  $\beta$ . For one thing the equations we derived are no longer valid but more importantly the increase in power is no longer dependent on delayed neutrons if  $\Delta \mathbf{k} > \beta$ . Return to our numerical example with a value of  $\Delta \mathbf{k} = .15$  with  $\beta = .1$ .

100 delayed 900 prompt 1000 x 1.15 x  $(1 - .1) \rightarrow 1040$  prompt

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1

As you see power is increasing without having to "wait" for the delayed neutrons. This shortens the reactor response time.

When  $\Delta k = \beta$  the reactor is critical on prompt neutrons alone, hence the name "prompt critical".

Figure 3 shows reactor period versus  $\Delta k$  for a reactor with a prompt lifetime of .001 seconds (i.e. the Candu reactors).



Reactivity v Period

As you can see nothing radically happens when the reactor approaches prompt criticality, the chain reaction is simply becoming less dependent on the delayed neutrons, hence power is changing more rapidly. In this regard we use the avoidance of prompt criticality as a design limit. (SDS1 trips the reactor at a period of 10 s and SDS2 trips the reactor at a period of 4 s. Both of these are below prompt criticality,  $T \approx 1$  s).

Also included on figure (3) is the plot of period versus reactivity for a reactor with only Pu-239 ( $\beta = 0.0021$ ) as a fuel. You will note that SDS1 and SDS2 trip set points provide adequate protection even in this situation. As we approach equilibrium fuel in our reactors we get closer to this situation. At equilibrium fuel prompt criticality occurs at  $\Delta k \approx 0.0035$ . The practical consequence of this is that the reactor regulating and protection system design must be based on the worst case which is equilibrium fuel.

### Large Negative Reactivities (Reactor Trips)

The equations developed for the prompt jump are equally valid for any insertion of negative reactivity except that you have a prompt drop followed by a stable negative period. Recalling that:

$$T = \frac{\beta - \Delta \mathbf{k}}{\lambda \Delta \mathbf{k}}$$

if:

 $|\Delta \mathbf{k}| >> |\beta|$  $T \approx -\frac{1}{\lambda}$ 

Thus the stable reactor period will be determined by the decay constant of the delayed neutron precursors. In fact it will be determined by the longest lived group of precursors, thus, the shortest reactor period possible after the prompt drop will be -80 s. In our reactors we have a very significant production of neutrons from the photoneutron reaction with deuterium thus the actual period will be somewhat longer.

#### ASSIGNMENT

- 1. Define reactor period (T).
- 2. Write the expression for reactor period considering both prompt and delayed neutrons.
- 3. Explain physically the way in which delayed neutrons effect the time response of neutron power.

- 4. Calculate reactor power 10 seconds after a step insertion of +2 mk of reactivity for fresh fuel ( $\beta$  = 0.0065) and for equilibrium fuel ( $\beta$  = 0.0035). P<sub>O</sub> = 50% and  $\lambda$  = 0.1 s<sup>-1</sup>.
- 5. What do we measure on our reactors that is related to reactor period? What is the relationship?

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Nuclear Theory - Course 227

SOURCE NEUTRON EFFECTS

The source neutrons available in CANDU reactors are those from spontaneous fission and those from the photoneutron reaction with deuterium. As mentioned in lesson 227.00-2, spontaneous fission produces a neutron flux which is approximately  $10^{-12}$ % of the full power flux. The strength of the photoneutron source depends on the number and energy of the photons present. At significant power levels (>10%) the gamma flux is directly proportional to the power level; thus, the photoneutron reaction produces a photoneutron flux which is proportional to the total neutron flux present. At low power levels and particularly when shutdown, the strength of the photoneutron source depends on the inventory of fission products which produce the high energy (>2.2 MeV) gammas required for the photoneutron reaction. The longest lived relevant fission product decay chain has a half life of about 15 days; thus the photoneutron source persists for several weeks after shutdown, decreasing essentially exponen-tially from approximately 10<sup>-5</sup>% of full power one day after shutdown. As we shall see shortly the values given for the actual neutron fluxes in the reactor due to the photoneutron source do not include any fission multiplication of this source.

## The Effect of Neutron Sources on the Total Neutron Population

In a critical reactor with no neutron sources other than induced fission, the neutron population in the reactor remains constant from one generation to the next. In other words, neutron losses due to absorption and leakage exactly take care of the excess neutrons generated by fission that are not required to keep the chain reaction going.

Now imagine a neutron source emitting  $S_0$  neutrons in each neutron generation time to be inserted into the reactor, and let this reactor be subcritical with a value of k just less than 1.

The number of neutrons present at the end of the first generation will be  $S_0$ , because that is how many are emitted in that time. At the end of the second generation, these  $S_0$  will have become  $S_0$ k neutrons, and another  $S_0$  neutrons will have been added by the source, giving us a total of  $S_0 + S_0$ k. At the end of the third generation these  $S_0 + S_0$ k neutrons will have become  $(S_0 + S_0 k)$ k neutrons, and again another  $S_0$  neutrons will have been added by the source to give us a grand total of  $S_0 + S_0 k + S_0 k^2$ .

If you pursue this sort of argument indefinitely, you will appreciate that we are going to end up with a final neutron population,  $S_{\rm m}$ , given by

$$S_{\infty} = S_{0} + S_{0}k + S_{0}k^{2} + S_{0}k^{3} + S_{0}k^{4} + - - - -$$
$$= S_{0}(1 + k + k^{2} + k^{3} + k^{4} + - - - -)$$

With k less than one, the sum

$$(1 + k + k^{2} + k^{3} + k^{4} + - - - -) = \frac{1}{1-k}$$

We can therefore say that

$$S_{\infty} = \frac{S_0}{1-k}$$
(1)

You can see that expression seems to be quite a reasonable one, because you can say if  $S_{\infty}$  is our final population, it will become  $S_{\infty}k$  after one more generation. If k is less than 1, this means that  $S_{\infty} - S_{\infty}k$  neutrons have been removed and these are made up by  $S_0$  new ones emitted.

For example if  $S_{\infty} = 5000 \frac{\text{neutrons}}{\text{generation}}$  and k = .8, these

5000 neutrons will become  $kS_{\infty}$  or 4000 neutrons in the next generation and the source  $S_0$  neutrons per generation time will make up the cycle losses ( $S_{\infty} - kS_{\infty}$ ) of 1000 <u>neutrons</u>. Thus; generation

$$S_{0} = S_{\infty} - kS_{\infty}$$

$$S_{\infty} = \frac{S_{0}}{1-k}$$
(1)

By observing that;

$$1 - k = -\Delta k$$

we can rewrite equation (1) as;

$$S_{\infty} = -\frac{S_0}{\Delta k}$$
(2)

Equation (1) and (2) are equivalent and you may use whichever one is convenient.

- 2 -

or

While we have developed equations (1) and (2) in terms of  $S_{\infty}$  and  $S_0$  in neutrons per generation, the equations are equally valid in terms of power in watts or percent of full power as was shown in lesson 227.00-8.

#### Subcritical Multiplication

It is important to realize that even in a reactor that is well below critical (eg, -40 mk, typical of a reactor trip) the equilibrium source level  $(S_{\infty})$  is 25 times greater than the actual photoneutron source.

$$S_{\infty} = \frac{S_0}{1 - .96} = \frac{S_0}{.04} = 25 S_0$$

The factor  $(\frac{1}{1-k})$  can be called the *subcritical multiplication* factor. In the example above, the subcritical multiplication factor is 25. Thus, the indicated source level  $(S_{\infty})$  is 25 times the actual source level  $(S_0)$ . This means that fission is producing 25 times as many neutrons as the source. The amount of subcritical multiplication depends only on the value of k. For example, if we used only half of the shutoff rods used in the previous example, such that we had -20 mk:

$$S_{\infty} = \frac{1}{1 - .98} S_0 = 50 S_0$$

Now the subcritical multiplication factor is 50.

In a subcritical reactor without a neutron source the neutron population would totally collapse; however, when a source is present it is not the major constituent of the neutron population (provided k > 0.5).

## Calculation of k in a Subcritical Reactor

Suppose a reactor is shutdown with a constant indicated power of 2 x  $10^{-5}$ %. The operator inserts +1 mk by withdrawing an adjuster and power stabilizes at 3 x  $10^{-5}$ %. Find the original value of k.

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For the first case before the reactivity addition;

$$P_{\infty} = \frac{P_{0}}{1-k_{i}}$$

$$2 \times 10^{-5} = \frac{P_{0}}{1-k_{i}}$$

After the reactivity addition;

$$3 \times 10^{-5} \% = \frac{P_0}{1 - (k_i + .001)}$$

Since  $P_0$  can be assumed to be the same in both cases, the equations may be solved for  $k_i$ .

$$P_{0} = (1 - k_{i}) \times 2 \times 10^{-5}\%$$

$$P_{0} = [1 - (k_{i} + .001)] \times 3 \times 10^{-5}\%$$

$$2 \times 10^{-5}\%(1 - k_{i}) = 3 \times 10^{-5}\%(.999 - k_{i})$$

$$2 - 2 k_{i} = 2.997 - 3 k_{i}$$

$$k_{i} = .997$$

You can always find the value of k in a shutdown reactor by changing reactivity, noting the power before and after a change and doing a simple calculation.

## Time Considerations

In a subcritical reactor the power will increase to a new equilibrium value each time positive reactivity is added. The magnitude of the increase and the time it takes for power to stabilize will depend on the value of k. The closer k is to one the larger the power increase for a given reactivity increase, and the longer the time for power to stabilize. This is demonstrated in Figure 1 where we start with  $P_{\infty} = 1 \times 10^{-5}$ % and k = 0.90 and add reactivity in +10 mk steps allowing  $P_{\infty}$  to stabilize after each reactivity addition.

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Power increase in a subcritical reactor.

Justification for relative magnitudes of the power increases can be done in a straightforward manner using equation (1) or (2) and is left to the student. The time consideration is somewhat more obscure. Assume we start with a source  $S_0 = 1000 \frac{\text{neutrons}}{\text{generation}}$  at time zero and ask the question, "How long will it take to reach equilibrium  $(S_{\infty})$ ? Equilibrium will be obtained when the effect of the first group of  $S_0$ neutrons has totally disappeared (ie,  $S_0 k^N = 0$  or  $k^N = 0$ , where N is the number of neutron generations). For example, we will compare the number of generations required for the first group of source neutron to disappear with k = 0.8, 0.9, and 0.95. The tabulated values are  $S_0 k^N$ .

k N	1	2	3	^	Δ	30	Δ	62	Δ	127
0.80	800	640	512	V	v	1	V	-	V	-
0.90	900	810	729		_	42		1		-
0.95	950	903	857			215		42		1

As you see, with k = 0.8 it would take about 30 generations to reach equilibrium, with k = 0.9 about 62 generations, and with k = 0.95 about 127 generations. (Strictly of course it takes an infinite time, this example gets to within 0.1%.)

# Effect of Sources when $k \ge 1$

When the reactor is critical, equations (1) and (2) don't apply because they are based on the assumption that the series  $1 + k + k^2 + k^3 + k^4$  ----) has a finite sum. When constant, and S<sub>0</sub> new ones will be added every generation, so that the neutron population will then merely increase indefinitely and at a constant rate of S<sub>0</sub> neutrons for every generation.

This rate of increase is insignificant if the reactor power is greater than  $\sim 10^{-2}$ % and would generally be obscured by the automatic regulation of the reactor. In a supercritical reactor any effects of the sources may be ignored.

## ASSIGNMENT

- 1. Explain why the total neutron population in a subcritical reactor is significantly higher than the source level.
- 2. Define the subcritical multiplication factor.
- 3. Explain why it is possible to have a constant fission rate in a subcritical reactor.
- 4. Assuming  $S_0 = 1000 \frac{\text{neutrons}}{\text{generation}}$  and the average neutron lifetime is 0.1 seconds, how long will it take to reach equilibrium when k = 0.999? (You need a calculator for this problem.)

J.E. Crist A. Broughton Nuclear Theory - Course 227

POWER AND POWER MEASUREMENT

We tend to use the term "power" rather loosely and we need to have clear understanding of what "power" we are talking about. The power we have referred to most frequently in this course is *neutron power* which is equivalent to the fission rate. However, the actual output of the reactor is in the form of heat energy and we call the heat output *reactor thermal power*. Normally we calibrate our instruments such that 100% neutron power corresponds to 100% of the thermal power required from the reactor to provide the design heat input to the turbine cycle.\*

The "power" we normally rate the overall unit by is the gross electrical power output of the generator. By way of example, Pickering-A reactors produce 540 MW(e), gross generator output, for a thermal power from the reactor of 1652 MW(th) which corresponds to an average thermal neutron flux of

5.3 x 10<sup>13</sup> <u>neutron.cm</u>

Thermal Power and Neutron Power

Thermal power is generally measured by measuring the primary heat transport flow rate (m) and temperature change ( $\Delta T$ ) in selected coolant channels (called fully instrumented channels). Recall from Thermodynamics (325) that:

> $Q = \dot{m}C\Delta T$ Where: Q = thermal power (watts [thermal])  $\dot{m} = flow rate (kg/s)$ C = Specific Heat  $(\frac{J}{kg^{\circ}C})$  $\Delta T = (T_{out} - T_{in})$  for the channel (°C)

\*At some stations the DCC automatically calibrates neutron power to be equal to thermal power above ~10% full power. Neutron Power is measured either by ion chambers located external to the calandria or by in-core flux detectors.

Thermal power has the advantage of being the actual, useful power output of the reactor. The measurements have the disadvantages of having an excessive time lag between neutron power changes and detected thermal power changes (around 25s, see 330.3 Lesson 34-2) and a non-linear relationship with neutron power especially at low power levels. The importance of the time lag may be seen by calculating the neutron power change that would occur in the time before there is any detected change in the channel  $\Delta T$  (assume this to be about 5s). With an inserting of + lmk of reactivity at equilibrium fuel: using equation (5) from lesson 227.00-8,

$$\frac{P}{Po} = \frac{\beta}{\beta - \Delta k} e^{\frac{\lambda \Delta k}{\beta - \Delta k} t}$$

$$= \frac{.0035}{.0035-.001} e^{\frac{(.1)(001)}{035-.001}5}$$
  
= 1.4 e<sup>.2</sup>  
= 1.7

Neutron Power would increase by a factor of 1.7 before detected thermal power even started to change. It should be clear that thermal power measurement is incapable of protecting the reactor from a rapid increase of reactivity, in fact it is rather slow even for normal control.

The non-linearity between thermal power and neutron power is due principally to fission product decay heat. Approximately 7% of the total reactor thermal power is produced by the  $\beta,\gamma$  decay of the fission products. Thus in a reactor operating at 100% of rated thermal output, 7% of the thermal power is due to decay heat. Even if it were possible to instantaneously stop all fissioning (neutron power  $\approx$  0%), the thermal output would still be 7% of full power and would decay over a long period of time. Figure 1 is a graph of a typical rundown of neutron power and thermal power after a reactor trip. Note that after a minute the neutron power makes very little contribution to the thermal power.



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Figure l

A second source of non-linearity is the heat lost from the coolant channels to the moderator (eg,  $\sim 4$  MW[th] at BNGS-A). The amount of heat lost is a function of the temperature difference between the coolant and the moderator and is, therefore, relatively independent of the power.

A third source of non-linearity is the heat generated by fluid friction. About two-thirds of the pressure drop in the heat transport system occurs in the coolant channels. This means that about two-thirds of the heat input of the heat transport pumps shows up in the coolant channels (eg,  $\sim$ 13 MW[th] at BNGS-A). This depends only on coolant flow rate and is independent of reactor power level.

Because of these non-linearities we must recalibrate neutron power to thermal power if the power level is changed.

## Power Monitoring when Shutdown

As you might surmize from Figure 1, thermal power and neutron power are not proportional when power is <10%. To protect the reactor against criticality accidents we must therefore monitor neutron power, especially at low power levels.

Assume for a moment that we built a protective system which used thermal power as the control variable. The reactor is slightly subcritical ( $_{\rm k}$  = 0.999) and the neutron power is  $10^{-5}$ %. Since the response time of the  $\Delta T$  detector is about 25 seconds, we would expect thermal power to lag neutron power such that, about 25 seconds after neutron power reacted, 1% thermal power would indicate 1%. Now assume the reactor is inadvertantly made supercritical ( $_{\rm k}$  = 1.003). For equilibrium fuel, +3 mk gives a reactor period of  $\approx$ 2 seconds. Thus 22 seconds after the reactivity addition, neutron power will reach 1%,  $\approx$ 25 seconds after that, thermal power will reach 1% and begin to show a rapid rate of change. In those intervening 25 seconds neutron power will reach  $\approx$ 27,000%.

If this reactor had been monitored for neutron power and rate of change of neutron power, the excursion could have been terminated long before power reached 1%. (Typically SDS 1 trips at a reactor period of 10s and SDS 2 trips at a period of 4 s.)

Uses of Power Measurements

1. Thermal Power is used for calibration of total neutron power and as a continuous checking function for zone power.

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- 2. Neutron Power is used in two ways:
  - a) Linear Neutron Power (linear N) may be used for indication, protection (high power trip) and/or control in the range of 15% to 120% neutron power.
  - b) The logarithm of Neutron Power (log N) is normally used for indication and control in the range of  $10^{-5}$ % to 15% neutron power (although the meter goes to 100% and controls to 100% if linear N fails).
- 3. Rate of change of neutron power may again be used in two different manners:
  - a) Linear Rate is the rate of change of linear neutron power displayed as percentage change of full power per second (%FP/s). (Not always used.)
  - b) Rate Log is the rate of change of the logarithm of neutron power in percent of present power per second (%/s). Rate Log is normally used for protection against excessive rates of change of power and is the inverse of reactor period. Recall that in its simplest form power may be expressed as:

$$P = P_o e^{t/1}$$

Then the natural logarithm of power is:

$$Ln P = Ln P_0 + \frac{t}{T} Ln e$$
  
or  $Ln P = Ln P_0 + \frac{t}{T}$ 

The rate of change of the log of power is the derivative with respect to time, thus;

$$\frac{d}{dt} \begin{pmatrix} Ln & P \end{pmatrix} = \frac{d}{dt} \begin{pmatrix} Ln & P_0 \end{pmatrix} + \frac{d}{dt} \begin{pmatrix} t/T \end{pmatrix}$$

$$\frac{d}{dt} \begin{pmatrix} Ln & P \end{pmatrix} = \frac{1}{T}$$
Rate Log

A typical trip setpoint (SDS-1) is a Rate log of 10%/s which corresponds to a reactor period of 10s.

# Behaviour of Power After a Trip

Assume a Candu reactor is running at 100% full power, with equilibrium fuel, and a reactor trip inserts -40 mk of reactivity. Figure 2 shows the theoretical behaviour of neutron power and thermal power. The remainder of this section will explain why the powers behave in this manner.

## Neutron Power Rundown

We will divide the power rundown into three regions. In region I the prompt neutron population is rapidly collapsing. With K = 0.96 (ie, with typical shutdown reactivity of -40 mk) the original prompt neutron population would decrease by a factor of 0.96 each generation. In 100 generations it would be less than 2% of its original value.

With a prompt neutron lifetime of 0.001 s, this decrease would take 0.1 s. Because of the delayed and photoneutrons, the actual neutron power will, however, not drop quite this fast nor will it drop this far.

Just before the reactivity insertion, delayed neutrons made up 0.35%\*\* of the neutron population (ie, 99.65% of the fissions were caused by prompt neutrons, 0.35% were caused by delayed neutrons). Immediately after the insertion, if we assume the prompt neutrons disappear we have a source of neutron (0.35% of full power) in a subcritical reactor; thus we can use the equation for neutron power in a subcritical reactor:

$$P_{\infty} = -\frac{P_{0}}{\Delta k}$$

$$P_{\infty} = -\frac{0.35\%}{-0.040}$$

$$= 8.75\% \text{ of full power.}$$

\*  $n = n_0 k^n$ 

$$\frac{n}{n_0} = (0.96)^{100} = 0.017$$

\*\*The delayed neutron fraction ( $\beta$ ) for equilibrium fuel is 0.0035.

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Therefore, the delayed neutrons present at the time of the trip will not let the neutron power drop initially below  $\sim$ 9% as shown in Figure 2. (Remember the actual drop is determined by the value of  $\Delta k$  inserted and the value of  $\beta$ .)

A second way to reach this conclusion is to recall from Lesson 227.00-8 that neutron power may be approximately calculated using:

$$P(t) = P_0 - \frac{\beta}{\beta - \Delta k} e^{\frac{\lambda \Delta k}{\beta - \Delta k} t}$$

where:

$$\frac{\beta}{\beta - \Delta k}$$

is the power after the prompt drop, which in this case gives;

$$P(o) = \frac{0.0035}{0.0035 + 0.040} \times 100\% = 8.05\%$$

which is nearly the same answer we got before. Either method is an acceptable approximation.

As the reactor is subcritical, the equation for power in a subcritical reactor applies throughout regions II and III. In region II the source of neutrons is the decay of the delayed neutron precursors which were present prior to shutdown. This source decreases rapidly at first as the short-lived precursors decay and slows down until the longest-lived group ( $t\frac{1}{2} = 55$  s) controls the rate of power decrease.

As was pointed out in Lesson 227.00-8, the reactor period after a large insertion of negative reactivity may be approximated as:

$$T \approx -\frac{1}{\lambda}$$

where,  $\lambda$  is the decay constant for the delayed neutron precursors.

The division between regions II and III is somewhat arbitrary. As the longer-lived delayed neutron precursors decay away, the photoneutrons are now the only important source of neutrons. Somewhere around 20 minutes after shutdown, the photoneutrons become the controlling source. From then on the power decreases at a rate determined by the decay of the fission fragments producing the 2.2 MeV photons required for the photoneutron reaction. As the longest-lived photoneutron producing fission fragments have half-lives of ~15 days, this source takes about 3 months to reduce to  $10^{-5}$ % full power.
## Thermal Power Rundown

At full power  $\sim7\%$  of the total thermal power is produced by the decay heat of the fission products (see Lesson 227.00-2). Although the fission rate can decrease very rapidly, the heat produced by decay of fission products (called decay heat) will only decrease at the decay rate of the fission products. Fission products have half-lives ranging from fractions of a second to thousands of years. Thus we expect a very slow decrease in thermal power. Typically thermal power will take about a day to decrease to 1% of full power. (For a Bruce reactor this is  $\sim29$  MW[th]).

The actual thermal power rundown will depend on the fission product inventory. A reactor at equilibrium fuel will have more fission products than one with relatively fresh fuel. therefore, it would produce a greater decay heat. This difference in production of decay heat will become more pronounced as time passes and the longer-lived fission products become more significant.

#### ASSIGNMENT

- 1. Discuss the advantages and disadvantages of neutron power and thermal power for controlling a reactor when:
  - a) At significant power levels (>10%)
  - b) When shutdown.
- 2. A reactor has been operating at 100% thermal and neutron power for a long time. Neutron power is reduced to 50%. Will thermal power be higher than, lower than, or equal to 50%? Explain your answer. (Assume calibration is done only at 100%.)
- 3. A reactor is operating at 15% thermal and neutron power. Neutron power is raised to 50%. Will thermal power be equal to, greater than, or less than 50%? Explain your answer. (Assume calibration is done only at 15%.)

- 4. A reactor is being started up by removing Boron from the moderator. Assume the ion exchangers (IX) remove the Boron at a constant rate. The power at one time on the He-3 counter is  $10^{-6}$ %. After one hour of IX removal, power stabilizes at 1.2 x  $10^{-6}$ %. How much longer will ion exchange be required before the reactor is critical?
- 5. Calculate the power after the initial drop in power if a trip inserts -30 mk in a reactor with fresh fuel ( $\beta$  = 0.0065). Use two methods.
- 6. Explain why, for a given reactor, the decay heat rate should be higher when it has reached equilibrium fuel than when it was running on fresh fuel.

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FISSION PRODUCT POISONING

All Fission products can be classified as reactor poisons because they all absorb neutrons to some extent. Most simply buildup slowly as the fuel burns up and are accounted for as a long term reactivity effect (as we did in lesson 227.00-7). However, two of the fission products, Xe-135 and Sm-149, are significant by themselves due to their absorption cross section and high production as fission products or fission product daughters. Xenon-135 has a microscopic absorption cross section of  $3.5 \times 10^6$  barns and a total fission product yield of 6.6%. Samarium-149 has an absorption cross section of 42,000 barns and a total fission product yield of 1.4%. Xenon-135 is the more important of the two and will be dealt with in more detail.

#### Xenon-135

Xenon-135 (often carelessly referred to just as xenon) is produced in the fuel in two ways:

- a) Directly from fission. About 0.3% of all fission products are Xe-135.
- b) Indirectly from the decay of iodine-135, which is either produced as a fission product or from the decay of the fission product tellurium-135 via the following decay chain:

 $5_2 Te^{135} \frac{\beta_{,\gamma}}{t_{2}^{1} = 30s} \quad 5_3 I^{135} \quad \frac{\beta_{,\gamma}}{t_{2}^{1} = 6.7h} \quad 5_4 Xe^{135}$ 

Te-135 and I-135 together constitute about 6.3% of all fission products. Due to the short half-life of Te-135 we normally consider the whole 6.3% to be produced as I-135. The rate of production of xenon and iodine from fission depends on the fission rate. Thus:

Rate of production of Xe-135 from fission =  $\gamma_{Xe} \Sigma_{f} \phi$ 

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Rate of production  $= \gamma_{I} \Sigma_{f} \phi$ of I -135 from fission  $= \gamma_{I} \Sigma_{f} \phi$ 

where:  $\gamma_{Xe}$  = Fission product yield of Xe

 $\gamma_{T}$  = Fission product yield of I

 $\Sigma_{f}$  = Microscopic fission cross section

 $\phi$  = Average thermal neutron flux

The rate of production of xenon from iodine depends only on the decay of the iodine, thus:

Rate of production of Xe -135 from I -135 =  $\lambda_{I}N_{I}$ 

where:  $\lambda_{I} = \text{decay constant for I } -135(S^{-1})$ 

$$N_{I} = \text{concentration of I} -135 \left(\frac{\text{atoms}}{\text{cm}^{3}}\right)$$

a) Radioactive decay as follows:

$$5 + Xe^{135} \xrightarrow{\beta \gamma} 5 5Cs^{135}$$

b) Neutron absorption (burnout):

 $_{5 4}Xe^{135} + _{0}n^{1} \longrightarrow _{5 4}Xe^{136} + \gamma$ 

Niether Cs -135 nor Xe -136 are significant neutron absorbers. The removal rates are as follows:

> Rate of change of Xe -135 by decay =  $\lambda_{Xe}N_{Xe}$

Rate of change of  $xe_{a} xe_{Xe^{\varphi}}$ 

where:  $\lambda_{Xe}$  = decay constant for Xe -135

 $N_{Xe}$  = Concentration of Xe -135  $\sigma_a^{Xe}$  = microscopic absorption cross section  $\phi$  = thermal neutron flux

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Now we can set up two equations, one which describes the behaviour of xenon and one which describes the behaviour of iodine. The time rate of change of the iodine  $(\frac{d}{dt}N_{I})$  is:

$$\frac{d}{dt} N_{I} = \underbrace{\gamma_{I} \Sigma_{f}}{\gamma_{I} \Sigma_{f}} - \underbrace{\lambda_{I} N_{I}}{\gamma_{I} \Sigma_{f}}$$
(1)

Production Loss due from fission to decay

The time rate of changes of the xenon  $(\frac{d}{d+} N_{xe})$  is:



We would like to examine the buildup of xenon in the reactor; however, since much of the xenon comes from iodine we must examine the behaviour of iodine first.

Examining equation (1) you can see that if we startup a reactor with no iodine present we will initially have a production term  $(\gamma_I \Sigma_f \phi)$  but no loss term since  $N_I$  is zero. As iodine is created the loss term grows ( $N_I$  is increasing) while production term remains constant (for constant power). Eventually the loss will equal the production and the iodine level will remain constant. Mathematically:

 $\frac{d}{dt}N_{I} = 0 = \gamma_{I}\Sigma_{f}\phi - \lambda_{I}N_{I}$ 

 $\lambda_{\mathbf{I}} \mathbf{N}_{\mathbf{I}} = \gamma_{\mathbf{I}} \Sigma_{\mathbf{f}} \phi$ 

$$N_{I} = \frac{\gamma_{I} \Sigma_{f} \phi}{\lambda_{I}}$$
  
Equili-

brium iodine concentration. This is a simple exponential buildup which can be considered to reach equilibrium after about five half-lives or 30 hours (within 3%). The buildup is shown in Figure 1.



Figure 1

Now we are able to examine the behaviour of xenon. The buildup of xenon is somewhat more complex than the buildup of iodine. Again at equilibrium,  $\frac{d}{dt} N_{Xe} = 0$ , and production equals loss. Also iodine must be in equilibrium so  $\gamma_I \Sigma_f \phi = \lambda_I N_I$ . Thus we can write:

$$\gamma_{Xe} \Sigma_{f} \phi + \gamma_{I} \Sigma_{f} \phi = \lambda_{Xe} N_{Xe} + q_{a} N_{Xe} \phi$$
(3)

Rearranging:

 $\underbrace{N_{Xe}}_{\text{Equili-}} = \frac{(\gamma_{Xe} + \gamma_{I}) \Sigma_{f} \phi}{\lambda_{Xe} + \sigma_{a}^{Xe} \phi}$ Equili-  $\lambda_{Xe} + \sigma_{a}^{Xe} \phi$ brium Xe concentration

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The buildup is shown graphically in Figure 2.

Figure 2

It again takes about 5 half-lives to reach equilibrium and for xenon this is about 50 hours.

It is useful to know the relative importance of the production and loss terms for xenon at equilibrium. Examining equation (3) we see the relative importance of the production terms depend only on their respective fission product yields. Thus direct production of Xe-135 from fission is about 5% of the total production at equilibrium while indirect production from the decay of I-135 is 95% of the total production.

In examining the loss terms, note that the loss due to decay depends only on the decay constant  $(\lambda_{Xe})$ . The loss due to burnout depends on the cross section  $(\sigma_a^{Xe})$  and the neutron flux. Therefore, the relative importance of the loss terms varies from reactor to reactor depending on the normal flux levels. For a given reactor the relative importance varies with power level. For our larger reactors (Bruce and Pickering) full power flux is  $7 \times 10^{13} \frac{n-cm}{cm^3 s}$ 

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Thus;  $\gamma_{Xe} = 2.1 \times 10^{-5} \text{ s}^{-1}$ 

$$\sigma_a^{Xe} = 24.5 \times 10^{-5} s^{-1}$$

Therefore, burnout constitutes more than 90% of the loss at full power.

### Reactivity

The reactivity worth of xenon (called Xenon Load) is a function of the concentration of xenon. As it is the reactivity due to xenon that we are concerned about, it is normal to express Xenon Load in reactivity units  $(\Delta k_{Xe})$ . As shown in Figure 3, the equilibrium Xenon Load for 100% is about -28 mk.

It is also common practice to express the concentration of iodine as Iodine Load in mk. It is important to realize that iodine is not itself a poison hence there is no actual reactivity associated with it. *Iodine Load* is by definition the reactivity if all the iodine present were instantaneously changed to xenon. I repeat it is not an actual reactivity.

By examining the equations for equilibrium xenon and iodine it can be deduced that equilibrium Iodine Load is a direct function of power (eg, doubling the power doubles the Iodine Load) whereas equilibrium Xenon Load does not have such a straightforward relationship with power. Figure 3 shows the approximate variation of equilibrium Xenon Load with power for Bruce or Pickering. The significant point is that equilibrium Xenon Load doesn't change much over the normal operating range.



#### Figure 3

## Xenon Simulation

The reactivity due to Equilibrium Xenon is easily compensated for by designing the reactor to have sufficient excess positive reactivity to overcome the negative reactivity due to the xenon. Now the regulating system must be capable of controlling the excess positive reactivity when there is no xenon present (eg, startup after a long shutdown). This is most commonly done by dissolving a poison (boron or gadolinium) in the moderator and removing it as the xenon builds up. This addition of poison to the moderator on startup is called Xenon Simulation.

As you may suspect, the buildup to and presence of Equilibrium Xenon does not present a significant problem in the operation of our reactors. However, the transient behaviour of xenon creates a major obstacle to operation.

#### Transient Xenon Behaviour

Assume a reactor has been operating at 100% power long enough for xenon to have reached equilibrium. If power is rapidly reduced to essentially 0%, what happens to the xenon concentration? To answer this question we shall examine the differential equation which describes the time behaviour of xenon.

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graphically in Figure 4.

$$\frac{d}{dt}N_{xe} = \gamma_{xe}\Sigma_{f}\phi + \gamma_{I}N_{I} - \gamma_{xe}N_{xe} - \sigma_{a} \qquad Xe_{N}_{xe}\phi$$
5% 95% 10% 90%

The percentages shown are the relative magnitudes of the production and loss terms prior to the decrease in power. When power is reduced to 0%, the small production term  $(\gamma_{Xe}{}^{\Sigma}f^{\phi} - direct fission production)$  and the large loss term  $(\sigma_{a}{}^{Xe}N_{Xe}^{\phi} - burnup)$  both cease. Since the major production term  $(\lambda_{I}N_{I} - decay \ of \ iodine)$  continues the concentration of xenon starts to increase. The increase can't go on forever since there is a limited supply of iodine, thus the xenon peaks and eventually decays away. This is shown



Figure 5

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Time to Poison Versus Size of Step Reduction in Power from an Initial Power Level of 200 MWe. Values for Other Initial Power Levels are Shown in Tabular Form. Fuel Assumed to be at Equilibrium Irradiation.

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40 OWABLE MAXIMUM LOAD TO AVOID POISONING OUT AL 38 INITIAL POWER (35<sup>°</sup> (7-1'') (WILLI, K REACTIVIT EXCESS APPROX. EQUIL IBRIUM LOAE FOR 200 ₩₩e 26 25 24 23 22 21 SIZE OF STEP REDUCTION IN POWER (MW) 20 Q 10 20 30 40 50 60 70 80 90

Maximum Xe load attained during the transient following step reductions in power from various initial power levels. (Xenon Assumed to be at the Corresponding Steady State Value Initially; Fuel Assumed to be at Equilibrium Irradiation).

Figure 7

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removal by neutron capture will also decrease by 40% from its full power value, but because xenon is still being removed the transient will not reach its shutdown peak. Looking at the figure, you will see that for a 40% reduction the available excess reactivity of ~10 mk is just sufficient to override the transient altogether. Ultimately equilibrium will be restored and the xenon load will then be that corresponding to 60% of the full power flux. The figure also shows that the <u>rate</u> of xenon build-up is less for a 60% reduction than for a 100% reduction, and that the poison override time would therefore be longer.

- Fig. 6 shows that this is true, namely that for a fixed amount of excess reactivity the poison override time depends on the size of the power reduction. For example, the curve shows that for a reduction of 120 MWe this time will be 1 hour, but it will be twice that for a reduction of 100 MWe.
- Fig. 7 shows the maximum xenon loads reached during the transient following step reductions from various initial power levels. For example, if the reactor is operating at 160 MWe and is then taken down to 100 MWe, the xenon load will increase from 27.2 mk to 35.1 mk. With 10 mk excess reactivity there should be no problem, but without looking at curves like this you wouldn't know whether there would be.

The converse to these curves also applies. For example, if the reactor is running at 140 MWe (at equilibrium) and it is taken to 200 MWe, the immediate effect will be a gain in reactivity due to increased burnup of xenon. At the same time more iodine will be produced which will not show up as extra xenon production until later on. As a result, the curve will run through a minimum, and than the xenon production will increase because of the increasing amount of iodine that is decaying. Eventually, the xenon concentration will attain the new equilibrium value corresponding to operation at 200 MW. The whole process is shown schematically in Fig. 8, and it does not normally present any operational problems.

#### Xenon Oscillations

So far, we have assumed that the xenon poisoning and reactivity loads apply to the reactor as a whole. No mention has been made of the possibility of localized changes in xenon poisoning which can have a very important effect on reactor stability.



Xenon Transient Following Step Load Increase From Various Initial Powers to 100% Final Power.

## Figure 8

For example, let us consider a reactor that has been operated at power long enough for the iodine and xenon concentrations in the fuel to have reached equilibrium.

Suppose now that without changing the total power of the reactor, the flux is increased in one region of the reactor and simultaneously decreased in another region. This change from the desired normal distribution is called a *flux tilt*. This may happen, for example, if control rods or similar mechanisms are inserted into one region and at the same time withdrawn from another. In the region of increased flux, the xenon now burns out more rapidly than it did prior to the change, and its concentration decreases. This decrease in xenon concentration leads to a higher reactivity in this region, which, in turn, leads to an increased flux. This again leads to increased local xenon burnup, increased local reactivity, increased flux, and so on.

Meanwhile, in the region of decreased flux, the xenon concentration increases due to its reduced burnup and to the continued decay of the existing iodine which was produced in the original, higher flux. This increased xenon concentration decreases the reactivity in this region, which reduces the flux, in turn, increasing the xenon concentration, and so on. The thermal flux, and hence the power density, thus decreases in this region while it increases in the other, the total power of the reactor remaining constant. These local power excursions do not continue forever. In the region of increased flux, the production of xenon from the decay of iodine, which is now being formed more rapidly in this region, ultimately reduces the reactivity there and the flux and power eventually decrease. Likewise, in the region of reduced flux, the accumulated xenon eventually decays, increasing the local reactivity and reversing the flux and power transient in that region.

In this way, the flux and power of a reactor may oscillate between different regions (end to end or side to side) unless action is taken to control them. Calculations, fortunately too lengthy to be spewed out here, show that these *xenon oscillations* have a period of from about 15 to 30 hours.

Xenon oscillations can only occur in large reactors. The argument to show this is as follows:

If the neutrons produced in one region of the reactor do not cause significant fissions in another region, then the two regions can act independently of one another. The criterion that determines whether or not this is possible is the degree of neutron leakage from the one region to the other. In a reactor such as NPD the core is small enough to permit a disturbance started in one region to have an effect in another region. The xenon and flux changes would therefore affect the whole core and a regulating system based on flux measuremen in one locality can correct the flux disturbance and prevent xenon oscillations from being initiated.

If the reactor is large, leakage of neutrons between regions is very small. A disturbance started in one region has little effect in another region. Thus, if a flux increase occurs due to a fuel change in one region, for example, a nonregional regulating system would compensate for this and maintain steady power by lowering the flux in another region to keep the average flux across the core constant. This would set up a xenon oscillation in the second region exactly out of phase with that in the first region.

Furthermore, it is obvious that xenon oscillations can only occur if the flux is high enough for xenon burnup to be as pronounced as xenon decay.

These two conditions for the presence of xenon oscillations (ie, large reactor size and high flux) are satisfied for most power reactors. Since xenon oscillations can occur at constant power they may go unnoticed unless the flux and/or power density distributions are monitored at several points in the reactor. This must be done in order to prevent such oscillations, since they represent something of a hazard to the safe operation of a reactor. Conceivably, they may lead to dangerously high local temperatures and even to fuel meltdown.

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In any event, these oscillations, if permitted to continue, burden the core materials with unnecessary temperature cycling which may result in premature materials failure.

One of the purposes of the regional absorber rods at Douglas Point, and of the regional liquid zone control systems at Pickering and Bruce, is to prevent such xenon oscillations. For example, at Pickering the reactors are subdivided into 14 regions (called zones), and each region has flux detectors whose output is used to adjust the amount of light water absorber in the zone control compartments.

#### Samarium-149

Sm-149 is the most important of the stable fission products. It is formed in the fuel by the decay of fission product neodymium-149 and promethium-149:

$$Nd^{1+9} \xrightarrow{\beta^{-}} Pm^{1+9} \xrightarrow{\beta^{-}} Sm^{1+9}$$

Since Sm-149 is stable, the only removal process for it is neutron capture. The Sm-150 formed has a low absorption and is therefore insignificant. Sm-149 has a much lower cross section  $(4.2 \times 10^{4} \text{b})$  than Xe-135, it will take correspondingly longer for equilibrium to be reached. The half-life of neodymium is so short compared to promethium that we lump its fission product yield with promethium. Note that there is no direct production of samarium from fission. As with xenon we need two equations to describe the behaviour:

dt N <sub>Pm</sub>	$= \gamma_{\mathbf{Pm}} \Sigma_{\mathbf{f}} \phi -$	$\overset{\lambda_{\mathtt{Pm}} \mathtt{N}_{\mathtt{Pm}}}{}$
	Production from fission	Loss due to decay

$\frac{d}{dt}$ <sup>N</sup> Sm	$= \underbrace{{}^{\lambda} Pm^{N}Pm}_{}$	$\sigma a Sm Sm^{\phi}$
	Production from decay fo Pm	Loss due to burnup

where: 
$$\gamma_{Pm}$$
 = Fission Product Yield of Promethium  
 $\Sigma_{f}$  = Fission Cross Section of the Fuel  
 $\phi$  = Average Neutron Flux  
 $\lambda_{Pm}$  = Decay Constant of Promethium  
 $N_{Pm}$  = Number Density of Promethium  
 $N_{Sm}$  = Number Density of Samarium  
 $\sigma_{a}^{Sm}$  = Cross Section of Samarium

The equilibrium Pm-149 concentration is:

$$N_{Pm} = \frac{\gamma_{Pm} \Sigma_{f} \phi}{\lambda_{Pm}}$$

Just as with iodine-135, the equilibrium concentration of Pm-149 is a direction function of the power level.

The equilibrium concentration of Sm-149 is:

$$N_{Sm} = \frac{\gamma_{Pm} \Sigma_{f}}{\sigma_{Sm}}$$

Note that equilibrium samarium is independent of the flux level. Equilibrium Samarium Load is around -5.5 mk and it takes about 300 hours of operation to reach equilibrium for our reactors (time to reach equilibrium is a function of the flux level).

## Samarium Growth After Shutdown

After a shutdown the samarium concentration will increase since none is being burned out and some is still being produced by the decay of Promethium. The maximum Samarium Load after shutdown depends on the promethium concentration prior to shutdown. For our larger reactors the maximum Samarium Load is about 12 mk. The buildup is shown in Figure 9.



It is interesting to note that although the equilibrium samarium load has to be allowed for in reactor design, the shutdown load may be ignored. There are two reasons for this.

- (1) By looking at the time scale of Fig. 9 you will realize that the maximum samarium load will not appear until the xenon transient has long been and gone. There will therefore be lots of reactivity available. You can also see that the increase in samarium load during the xenon poison override time is negligible, so that this doesn't present a problem either.
- (2) The rate at which the samarium is formed is governed by the Pm-149 half-life of 53 hours, and it corresponds almost exactly to the rate at which Pu-239 is formed after a shutdown. (Pu-239 is still produced from Np-239 decay, but it is not being used up since there are no neutrons). It turns out that the increased reactivity from this plutonium transient more than compensates for the increased samarium load. The net change after shutdown is about +6 mk due to combined effect of samarium and plutonium.

## ASSIGNMENT

- 1. Write the equations for the time rate of change of  $N_{Xe}$  and  $N_{I}$ . Explain what each term represents and give the magnitudes of the terms. Note the conditions under which these magnitudes are applicable.
- 2. Explain why equilibrium Xenon Load changes very little when power is raised from 50% to 100%.
- Explain why peak xenon after shutdown from 100% equilibrium will be nearly twice what it is after shutdown from 50% equilibrium.
- 4. Give and explain the conditions required for a xenon oscillation to occur.
- 5. Define Iodine load and explain its significance.
- 6. Explain why samarium growth after shutdown may be neglected in reactor design.

J.U. Burnham J.E. Crist

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#### Nuclear Theory - Course 227

REACTIVITY EFFECTS DUE TO TEMPERATURE CHANGES

In the lesson on reactor kinetics we ignored any variations in reactivity due to changes in power. As we saw in the previous lesson there are marked changes in reactivity due to xenon; occurring over a period of minutes to hours after an overall power change. Changes in reactor power causes changes in the temperature of the fuel, moderator, and coolant. These also have an effect on reactivity which is more rapid than xenon effects.

## The NRX Experiment

In 1949, the NRX reactor at AECL, Chalk River, was allowed to "run away". NRX is a heavy water moderated reactor which uses control rods for reactor regulation. The heavy water level was set 3 cm above the height at which the reactor would be critical at low power with the rods withdrawn. The reactor power was allowed to increase unchecked, and the manner in which it increased is rather unexpected (see Figure 1).

The power initially increased exponentially with a period of 33 seconds (T = 33 s, $\Delta \mathbf{k}$  = +1.6 mk). However, it did not increase indefinitely as you might have expected. As the temperature of the fuel rods increased, the reactivity decreased and this caused the rate of power increase to slow down. Later the reactivity decreased at a faster rate as the heavy water got warmer. The total decrease in reactivity was enough to make the reactor subcritical, and the end result was that the power reached a maximum value and then started to decrease.

Thus the reactor is self-regulating with temperature increases preventing the power from continuing to increase. Of course, in this experiment the initial excess reactivity was quite small; if more reactivity had been inserted initially it is quite possible that the power would have continued to rise. The point of this example is not to demonstrate that reactor power would never increase continuously (it well might), but to show that there was a loss in reactivity due to the increase in the temperatures of fuel and heavy water.



Fig. 1 The NRX Experiment

The temperature coefficient of reactivity is defined as the change in reactivity per unit increase in temperature. Its units are mk/°C.

The coefficient may be positive or negative. In the example just described it was negative, because an increase in temperature led to a loss or reactivity.

Temperature changes occur, more or less independently, in the fuel, the heat transport system and the moderator, and there will therefore be a temperature coefficient of reactivity associated with each of these. It is very desirable for the overall temperature coefficient of a reactor to be negative to provide the self-regulating feature illustrated by NRX.

In order to fully understand why changes in temperature cause changes in reactivity it is necessary to understand both the physical and nuclear properties which change with temperature.

## (a) Thermal Expansion Effect

As the temperature of the coolant and/or moderator increases its density decreases. As a result neutrons travel further thus, they have an increased probability of escaping ( $\Lambda_f$  and  $\Lambda_{th}$  may both decrease). Also with fewer moderator molecules there is less absorption in the moderator and thermal utilization (f) increases.

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## (b) Direct Nuclear Effect

This is the effect commonly known as Doppler Broadening. We mentioned earlier in the course that resonance capture occurs in U-238 for certain neutron energies related to the target nucleus which was assumed to be at rest. The resonance is actually determined by the relative velocity of the neutrons and the target nuclei. When the fuel gets hot, the uranium atoms will vibrate more vigorously. А neutron which would have been outside the resonance peak if the uranium atoms had been at rest, may encounter an atom moving at the necessary speed to put their relative velocity in the resonance peak. Thus the neutron, which might have survived in cold fuel, is now captured in hot fuel, and this is reflected in a spreading of the resonance peak as shown in Figure 2. There will then be a decrease in the resonance escape probability p and in the reactivity due to this so-called Doppler Broadening of the resonance peak\*.



Fig. 2 Doppler Broadening

\*Without a rigorous mathematic treatment it may not be easy to convince you that although the area under the curve is the same, the absorption increases. A simple (but basically correct) approach is to say that although  $\sigma_a$  for hot fuel is only half of what it is for cold fuel, it is high enough to virtually guarantee absorption of any resonance energy neutrons entering the fuel. Only now the resonance energy range has been doubled.

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## (c) Indirect Nuclear Effect

A thermal neutron is one which is in thermal equilibrium with its surroundings. Clearly then any change in the temperature of the moderator, coolant, or fuel will affect the average thermal neutron energy. Thus neutron cross sections, being energy dependent, are affected. This may affect the thermal utilization (f) and the reproduction factor (n). Generally the changes in n which are most significant, are due to changes in the ratio of the fission cross section to the absorption cross section of the fissile material  $(\sigma_f/\sigma_a)$ .

Figure 3 shows the variation of  $\eta$  for  $U^{2\,3\,5}$  and  $Pu^{2\,3\,9}$ . Note in particular that around 0.3 eV,  $\eta$  for  $Pu^{2\,3\,9}$  starts to rise rapidly



To evaluate the magnitude of the effects mathematically the Design Manuals evaluate the derivative of  ${\bf k}$  with respect to temperature

dk dr

$$\mathbf{k} = \varepsilon \mathsf{pnf} \Lambda_{\mathsf{f}} \Lambda_{\mathsf{f}}$$

$$\frac{1}{k} \frac{d}{dT}k = \frac{1}{\varepsilon} \frac{d\varepsilon}{dT} + \frac{1}{P} \frac{dP}{dT} + \frac{1}{\eta} \frac{d\eta}{dT} + \frac{1}{f} \frac{df}{dT} + \frac{1}{\Lambda_f} \frac{d}{dT} \Lambda_f + \frac{1}{\Lambda_t} \frac{d}{dT} \Lambda_t$$

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The change in each of the factors is tabulated in Table I for both fresh and equilibrium fuel. We will now look at the temperature coefficients for the fuel, moderator and coolant.

#### Fuel Temperature Coefficient

There are two primary effects due to an increase in the fuel temperature:

- 1) Increased resonance absorption
- An altered ratio of fission to absorptions in 2) the fuel.

Let us look at a concrete example. Table I gives makeup of the fuel temperature coefficient for the Pickering units at nominal operating conditions.

From this table you can see that the predominant term is the resonance capture term. It is sufficiently large to ensure an overall negative fuel temperature reactivity effect at nominal operating conditions, and it therefore provides the self-regulating feature that is so desirable.

#### TABLE I

Fuel Temperature Coefficient For Pickering Units 1-4

(Nominal Operating Conditions. Units are  $\mu k/^{\circ}C$ )

	Fresh Fuel	Equilibrium Fuel
(1/ε)dε/dT	0	0
(1/p)dp/dT	-9.33	-9.29
(l/f)df/dT	-0.79	+0.34
(l/ŋ)dŋ/dT	-4.04	+5.33
$(1/\Lambda_{f}) d\Lambda_{f}/dT$	0	0
$(1/\Lambda_t) d\Lambda_t/dT$	-0.83	-0.43
TOTAL	-14.99	-4.05

The resonance escape term  $\begin{pmatrix} \frac{1}{P} & \frac{dP}{dT} \end{pmatrix}$ is negative

because increasing the fuel temperature causes increased resonance capture due to doppler broadening. Fresh and

equilibrium fuel values are the same because the amount of  $U^{2\,3\,8}$  in the reactor is essentially constant.

The reproduction factor term  $\left(\frac{1}{\eta}, \frac{d}{dT}, \eta\right)$  is negative for

fresh fuel because the fissile material is all  $U^{235}$  and  $\eta$  decrease with increasing temperature in the  $U^{235}$  for energies of interest (< 1 ev) as shown in Figure 3. For equilibrium fuel this term is positive due to the increased concentration of  $Pu^{239}$ . The increase in  $\eta$  with temperature for  $Pu^{239}$  overwhelms the negative effect of the uranium.

The behavior of the thermal utilization term is also due to the increased concentration of plutonium. (The plutonium increases at 80% of the uranium 235 depletion. Thus 0.8 x 741.6 = 593 b > 580 b the cross section for U235.)

The change in thermal leakage is due to an increase in the distance a thermal neutron diffuses, which is brought about by an overall reduction in the thermal absorption cross section of the whole core.

## Heat Transport Temperature Coefficient of Reactivity

The reactivity effect associated with a change in coolant temperature is rather more complicated in its make-up than the fuel temperature effect, and we won't discuss it in detail.

Figure 4 shows the overall coolant temperature coefficient of reactivity for the Pickering units as calculated from the design data. It is very difficult to determine it from measurements, because you can't change the coolant temperature without changing the fuel temperature. It is however positive.

Figure 5 shows the results of measurements made on Pickering Unit 3 when it contained fresh fuel. The heat transport system was heated by running the primary pumps while the reactor was held critical at 0.1% of full power. The measurements extended over a period of 13 hours so that one must assume that the fuel temperatures kept in step with the coolant temperatures. The measured changes in reactivity therefore reflected both the fuel and the heat transport coefficients of reactivity, and you can see that the negative effect of the former more than compensates for any positive effect of the latter. The reactivity change is seen to be -7 mk from cold shutdown to hot shutdown.

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Figure 5

### Moderator Temperature Coefficient of Reactivity

As with the fuel temperature coefficient there are two effects; change in moderator density and increasing average thermal neutron energy. The temperature of the moderator affects the neutron energy much more than coolant or fuel does - it is the base temperature, so to speak. One would therefore expect the magnitude of the moderator coefficient to be greater than the other two, and this is in fact the case, as you can see from Table II which again gives the values applicable to Pickering.

## TABLE II

# Moderator Temperature Coefficient for Pickering Units 1 - 4

	Fresh Fuel	Equilibrium Fuel
(1/ε)dε/dT	0	0
(l/p)dp/dT	-24.0	-23.9
(1/f)df/dT	55.4	67.1
(1/ŋ)dŋ/dT	-59.2	76.0
$(1/\Lambda_{\rm f}) d\Lambda_{\rm f}/dT$	-13.0	-13.0
$(1/\Lambda_t) d\Lambda_t/dT$	-28.7	-22.0
TOTAL	-69.5	+84.2

(In units of  $\mu k$ /°C, calculated for  $\Delta T = -13$ °C)

The change in moderator density is responsible for an increase in the distance a neutron travels in slowing down. This in turn leads to a decrease in the resonance escape probability, p, as well as in the fast non-leakage probability.

The distance a neutron diffuses also increase. It is not only affected by the change in moderator density, but also by the reduction in all the absorption cross sections with increasing thermal energy. Consequently, the change in thermal leakage is greater than that in fast leakage.

The great changes in the value of  $\eta$  from fresh to equilibrium fuel are due to the effects of the ratio of fission to absorption in Pu<sup>2 39</sup> and U<sup>2 35</sup> as previously stated.

The thermal utilization term is always positive due to a decrease in absorption by the moderator associated with a decrease in moderator density.

### Practical Aspects

We have already mentioned that it is desirable for the temperature coefficients to be negative so that a selfregulating feature is provided. However, more must be considered than just the values of the three temperature coefficients. Two most important additional factors are; the size of the various temperature changes for a given power change, and the time period over which the changes occur.

Typically, in a change from hot shutdown, to 100% power, the <u>average</u> coolant temperature may increase by  $\approx 20 - 40^{\circ}$ C while the <u>average</u> fuel temperature will increase by 500 to 600°C and the moderator temperature will be maintained constant. Furthermore, the fuel temperature will change nearly instantaneously as the power changes while the coolant temperature change will lag the power change by a few seconds.

Thus, we achieve the desired self-regulation merely by having a negative fuel temperature coefficient of reactivity.

A negative temperature coefficient does, however, create some problems. In heating the fuel and coolant from a cold shutdown condition to a hot shutdown condition there is a net loss of reactivity worth which can be as much as 9 mk. Also, when power is increased there is a reactivity loss which must be compensated for. In Ontario Hydro, this is expressed in terms of the *power coefficient*, which is defined as the reactivity change in raising power from hot shutdown to 100% full power. It only includes the temperature coefficients of reactivity, and not any reactivity loss due to fission product formation. It is typically of the order of 5 or 6 mk for a heavy water reactor.

#### Effects Due to Void Formation

Voids will be formed if either the moderator or the heat transport system fluid boils. Void formation in the coolant is of more concern than in the moderator, and so we'll restrict our discussion to the effects of loss of coolant.

Because the reactivity increases with loss of liquid coolant, knowledge of the magnitude of this effect is important for safety reasons. The liquid coolant may boil as a result of:

- rupture of the feeder pipe(s)
- failure of the primary pump(s)
- large power excursions
- channel blockage.

Under all these circumstances the coolant will gradually be displaced by steam, and eventually the channel(s) may become totally depleted of liquid coolant. This is frequently called voiding the channel.

The severity of the above emergency conditions depends primarily on the rate of reactivity addition, although the total reactivity addition may be of equal importance. For a light water cooled reactor, such as Gentilly, loss of coolant results in a very large change in reactivity. For example, it is estimated that for Gentilly, operating with fresh fuel, the reactivity change for a loss of coolant in half the core can be as high as 37 mk, depending on the operating conditions at the time. This colossal change is of course primarily due to the increase in the thermal utilization, f, caused by the loss of  $H_2O$  absorber.

For  $D_20$  cooled reactors, the effects are nowhere near as drastic, although they are still very important.

Voiding of fuel channel causes a decrease in the moderation of neutrons in the immediate neighborhood of the fuel elements. Looking at figure 6 (a quadrant of a fuel bundle) you can see that a neutron born in one fuel element (eg, element 'A') normally passes through some coolant before reaching the next fuel element (element 'B') with the coolant providing a little moderation. With the channel voided there is no moderation hence, higher energy neutrons are interacting with the fuel in element B.



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This has two effects which can be seen by looking at the radiative capture and fission cross sections of  $U^{238}$  shown in Figure 7



- (a) An increase in the fast fission factor ( $\epsilon$ ) since  $\sigma_{f}$  increases with increasing energy.
- (b) An increase in the resonance escape probability (p) since  $\sigma_{n,\gamma}$  decreases with increasing neutron energy.

Both of these give rise to a positive void coefficient.

Voiding of the coolant also reduces the amount of absorbing material in the reactor, however, for heavy water coolant, this decrease is very small provided the coolant isotopic is high. In practice there is a lower limit on coolant isotopic to prevent an exessively large void coefficient. This lower limit is usually defined in Station Operating Policy and Principles. (eg, 97% at Bruce NGS 'A').

Excessive positive or negative void coefficients are to be avoided if possible. An excessively large positive coefficient will cause large power surges, during the void formation, which are likely to cause severe damage to the reactor if the protective system does not respond enough.

Excessive negative coefficients, on the other hand, cause a rapid decrease in power when the void is formed, which is then corrected for by the regulating system. Then, when the void fills, a power surge again results.

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## ASSIGNMENT

- 1. Explain why the fuel temperature coefficient of reactivity is more important than either the coolant or moderator temperature coefficient of reactivity. (Two reasons.)
- 2. Explain why the fuel temperature coefficient is larger in magnitude for fresh fuel than it is for equilibrium fuel.
- 3. Cite an example of when the moderator temperature coefficient of reactivity may be useful.
- 4. Considering only the effect on the void coefficient, explain why it is undesirable to add soluble poison to the coolant.

J.E. Crist

Nuclear Theory - Course 227

REACTIVITY CONTROL

Reactivity mechanisms represent the final control elements which cause changes in the neutron multiplication constant k (or reactivity  $\Delta k$ ) hence, reactor power. There are two separate requirements of the reactivity mechanisms which are preferably fulfilled by two independent systems. These requirements are:

- 1. Reactor Regulation. The three basis functions of the reactor regulation systems are:
  - a) Maintain k = 1 for steady power operation.
  - b) Provide small changes +ve or -ve in  $\Delta k$  to change reactor power.
  - c) Prevent the development of flux oscillations.
- 2. Reactor Protection. The principal purpose of the protective system is to rapidly insert a large amount of negative reactivity to shutdown the reactor (TRIP).

From a reactor safety viewpoint it is desirable to have reactor regulation and protection performed by separate systems. From a practical viewpoint no single system can adequately fulfill all the requirements for reactor regulation let alone regulation and protection together.

#### Requirements of Reactivity Mechanisms

As well as independence between (1) and (2) the complex physical and nuclear changes occurring in core during reactor operation mean that an effective regulating system will have to consist of more than one type of reactivity mechanism. A convenient breakdown of the various in core reactivity changes which require compensating/regulating controls is listed in Table 1 and grouped in terms of the most important parameters of any reactivity mechanism namely:

(i) reactivity worth (or depth)  $\Delta k$  (mk).

This must be somewhat larger than the reactivity change for which the mechanism must compensate or control, and

(ii) operational time interval.

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This is the time period during which the mechanism has to be able to supply or remove reactivity and this will hence determine the reactivity insertion rate (sometimes called the ramp reactivity rate),  $\Delta k$  per unit time (mk/s).

Each of the tabulated reactivity changes is now briefly described and typical  $\Delta k$  worths necessary to adequately control these changes as they occur in our stations are shown for comparison in Table 2. Where these values change from fresh fuel to equilibrium fuel load conditions then the difference is noted.

## In Core Reactivity Changes

(a) Power Changes (Ref. Lesson 227.00-12)

Because the temperatures of the fuel and coolant increase as power increases from a hot shutdown condition to a hot full power condition, reactivity changes. Under normal (ie, non excursion) type conditions there will be a negative reactivity worth change called the <u>power coefficient of reactivity</u>. These are tabulated in Table 2. In order to maintain criticulity an equal but opposite reactivity worth must be supplied by some other means, (eg, by removing an equivalent reactivity worth from the Zone Control System).

(b) Fuel and Coolant Temperature Changes (Ref. Lesson 227.00-12)

As the fuel and coolant are heated from a cold shutdown condition ( $^{25^{\circ}}$  C) to a hot shutdown condition ( $^{276^{\circ}}$ C) reactivity decreases, Table 2.

### (c) Moderator Temperature Changes (Ref. Lesson 227.00-12)

Normally moderator temperature is kept fairly constant (typically 70°C maximum in the calandria and 40°C at the heat exchanger outlets) but variation could be obtained by changing the rate of heat removal from the heat exchangers. The accompanying reactivity change is usually negative with increasing temperature for a freshly loaded core but changes to a small positive value at equilibrium fuel burn up as shown in Table 2.

(d) Fresh Fuel Burn Up (Ref. Lesson 227.00-7)

From an initial fresh fuel charge to equilibrium fuel burn up there is a large increase in negative reactivity load over a period of 6 - 7 months as a result of build up of long lived neutron absorbing fission products (not including  $Xe^{135}$ ) and depletion of fissile material. Figures for our reactors are quoted in Table 2. This is a slow but continuous reactivity change.

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## (e) Equilibrium Fuel Burn Up

At equilibrium fuel burn up, when the operating target excess reactivity has been reached, fission products continue to be built up and fissile material continues to be depleted. Continuous on power refuelling is of course the most important method of compensating for this continual depletion of fissile material at equilibrium burn up. The rate of reactivity loss for our reactors without refuelling is shown in Table 2 and for comparison the reactivity increases due to the refuelling of a single typical central channel are also listed.

(f) Equilibrium Xe Load Build Up (Ref. Lesson 227.00-11)

Following a long reactor shutdown (>2 - 3 days) an equilibrium reactivity load (up to 28 mk see Table 2) will be built up due to  $Xe^{135}$  accumulating in the fuel after start up.

(g) Xe Transient Build Up (Ref. Lesson 227.00-11)

Within 12 hours of a reactor shutdown (or large derating due to operational problems, or a load following situation) there is a very large transient rise in Xe poison concentration (up to -80 mk above the equilibrium level at Pickering, Table 1). To enable us to restart the unit, Xe OVERRIDE or BOOSTING CAPABILITY is provided to compensate for this reactivity loading providing an override time, measured after shutdown, which gives reactivity capability of restarting a unit within this time. Actual reactivities available and the override times thus obtained are listed in Table 2 for all our stations.

(h) Flux Oscillations (Ref. Lesson 227.00-11)

As localized flux/power changes occur in the core (from, for example, refuelling part of a channel or movement of a localized control rod) these can result in quite large undamped power swings (Xenon oscillations) being set up with periods between 15 - 30 hours.

To counterbalance these oscillating unbalanced reactivity loads in various regions (called ZONES) of the core, the ZONE CONTROL system is used. Total reactivity worth of these systems are shown in Table 2, and are actually larger than required to control only the flux oscillations as these systems are also used for bulk power control.

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# (i) Plutonium and Samarium Build Up (Ref. Lessons 227.00-7&11)

After shutdown plutonium builds up from the decay of neptunium adding positive reactivity and samarium builds up from the decay of Promethium adding negative reactivity. The overall effect is positive as shown in Table 1.

## TABLE 1

## In core reactivity changes

	Source of in-core reactivity changes.	∆k depth	time interval
(a)	Power changes, hut shutdown to hot full power.	medium (+ve, -ve)	seconds, minutes
(b)	Fuel and Coolant temperature changes.	medium (+ve, -ve)	seconds, minutes
(c)	Moderator temperature change.	small (+ve, -ve)	minutes
(d)	Fresh fuel burn up.	large (-ve)	6 - 7 months
(e)	Equilibrium Xe load build up.	large (-ve)	40 hours
(f)	Xe transient build up.	large (-ve)	<12 hours
(g)	Flux Oscillations.	medium (+ve, -ve)	15 - 30 hours
(h)	Equilibrium fuel burn up.	small (-ve)	days (continuous)
(i)	Plutonium and Samarium build up.	medium (+ve)	300 hours

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| REACTIVITY WORTH CHANGE                      |                                  |                     | NPD         | DOUGLAS<br>POINT | PICKERING<br>A & B | BRUCE<br>A & B |
|--|----------------------------------|---------------------|-------------|------------------|--------------------|----------------|
| (a)  | Power Coefficient                | fresh fuel          | -3.3 mk     | -6 mk            | -7 mk              | -9 mk          |
|  | hot shutdown -<br>hot full power | equilibrium<br>fuel | -1.2 mk     | -5 mk            | -3 mk              | -3.5 mk        |
| (b)  | Fuel and Coolant                 | fresh fuel          | -3 mk       | -б mk            | -8 mk              | -9 mk          |
|  | temperature 25°C<br>to 275°C     | equilibrium<br>fuel | -l mk       | -4.5 mk          | -2.5 mk            | -3 mk          |
| (c)  | Moderator Temperature            | fresh fuel          | -0.08 mk/ C | -0.06 mk/ C      | -0.06 mk/ C        | -0.07 mk/ C    |
|  | Coefficient                      | equilibrium<br>fuel | +0.01 mk/ C | +0.03 mk/ C      | +0.08 mk/ C        | +0.09 mk/ C    |
| (d)  | Fresh Fuel Burn Up               | -9 mk               | -20 mk      | -26 mk           | -22 mk             |                |
| (e)  | Xe Equilibrium Load              |                     | -24 mk      | -28 mk           | -28 mk             | -28 mk         |
| (f)  | Xe Peak Load                     |                     | -46 mk      | -107 mk          | -98 mk             | -105 mk        |
|  | Xe Override Capabilit            | Y*                  | +2.4 mk     | +10 mk           | +18 mk             | +15 mk         |
|  | Xe Override Time                 |                     | 35 min      | 30 min           | 45 min             | 40 min         |
| (g)  | Zone Control Reactivi            | NONE                | 3 mk        | 5.4 mk           | 6 mk               |                |
| (h)  | Reactivity Loss (Equi            | -0.15 mk/day        | -0.3 mk/day | -0.3 mk/day      | -0.5 mk/day        |                |
| Reactivity Gain/Refuelled<br>Central Channel |                                  |                     | +0.1 mk     | +0.2 mk          | +0.2 mk            | +0.5 mk        |
| (i)  | Plutonium and Samariu            | m Build Up          | +2.5 mk     | +6 mk            | +6 mk              | +6 mk          |

TABLE 2: COMPARISON OF STATION REACTIVITY LOADS

\* New elements only, will decrease by ~30% at end of life burn up.

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As you can see the range of reactivity depths and insertion rates make it impractical to try to design a single control mechanism.

## Methods of Reactivity Control

Before we can discuss actual control mechanisms we must look at the theoretical methods of reactivity control. Recalling that:

$$\mathbf{k} = \epsilon p \eta f \Lambda_f \Lambda_f$$

we will examine which of the six factors we can use to change/ control reactivity (remember  $\Delta k = k - 1$ )

First neither the fast fission factor (
$$\varepsilon$$
) nor the resonance escape probability (p) are easily varied. They depend on the amount of U-238 present and the lattice spacing in the reactor. Therefore, we make no attempt to control reactivity by controlling  $\varepsilon$  or p.

Next is the reproduction factor (n).

Recall that:

$$\eta = v \frac{\sum_{f}^{fuel}}{\sum_{a}^{fuel}}$$

If we increase the amount of fissile material present  $(\Sigma_{f}^{fuel})$  we will increase  $\eta$ . That is, more neutrons will be produced per neutron absorbed by the fuel.

<u>Thermal utilization (f) is the fraction of neutrons</u> absorbed by the fuel to those absorbed in the whole core:

$$f = \frac{\sum_{a}^{fuel}}{\sum_{a}^{fuel} + \sum_{a}^{non-fuel}}$$

If we increase or decrease the amount of non-fuel absorption, we vary f, hence reactivity. Variation of neutron absorption is by far the most common method of control.

Finally we have the <u>fast and thermal non-leakage</u> probabilities  $(\Lambda_{f} \& \Lambda_{t})$ . If we vary the leakage of neutrons from the reactor we will vary reactivity.

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## Reactivity Mechanisms

In order to discuss the reactivity mechanisms presently in use we shall divide them into five groups based on their basic function in the reactor. The five functional groups are:

- Automatic Reactor Regulation (includes bulk power and zone control)
- 2) Xenon Override.
- 3) Long Term Reactivity Control (includes fresh fuel burn up, the build up of equilibrium xenon and the build up of plutonium and samarium after shutdown).
- 4) Equilibrium Fuel Burn up.
- 5) Shutdown Systems

For each of these catagories we will discuss the methods used and the significant advantages and disadvantages of those methods (See 433.50-1 for a discussion of the mechanics of the systems). Table 3 indicates which systems are used at each station and the reactivity depth of each system.

## Automatic Reactor Regulation

a) <u>Moderator Level Control</u>. Small changes in moderator level change the thickness of the reflector on top of the reactor thus varying leakage ( $\Lambda_{f} \in \Lambda_{+}$ ).

Advantages:

1) Easily incorporated into a system using moderator dump for protection.

Disadvantages:

- 1) Zone control is not possible.
- 2) Lowering the moderator level distorts the overall flux distribution.
- b) <u>Control Absorbers.</u>

Solid rods of a mildly absorbing material (typically stainless steel) which can be operated vertically in the core. Because they are parasitic absorbers the control absorbers change the thermal ulitization (f).

Advantages:

1) Provide additional reactivity at minimal cost.

Disadvantages:

- In core guide tubes represent, permanent, reactivity loss (fuel burn up loss).
- c) <u>Liquid Zone Control (LZC)</u> Zone Control Compartments inside reactor which contain a variable amount of light water (a mild neutron absorber). Varying the amount of light water in the LZC, varies parasitic absorption hence thermal utilization (f).

Advantages:

- 1) Individual zone levels can be independently varried for zone control.
- Operating equipment is mainly outside containment, therefore, accessible during reactor operation.
- 3) Cooling easily accomplished.
- 4) Only slight distortion of the overall flux pattern.

Disadvantages:

- 1) Requires special design to insure that the zones fail safe (ie, fill).
- In core structure represents a reactivity (or fuel burn up) loss.

# Xenon Override

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## a) <u>Booster Rods</u>.

Solid rods of highly enriched (~90%) U-235. Insertion of booster rods increases the amount of fissile material in the reactor hence the reproduction factor (n). It also increases f.

Advantages:

1) Can provide large override capability.

Disadvantages:

- Enriched Uranium is a very expensive, non-Canadian product.
- Require highly reliable source of cooling (loss of cooling to an inserted booster at high power could cause the rod to melt down in about 5 seconds).
- 3) Because of cooling requirements additional trips are required thus complicating the reactor protection systems.
- 4) Limited lifetime as the reactivity worth decreases with each use.
- 5) A criticality hazard exists in the storage of both new and irradiated booster rods.
- 6) Because of all of the above reasons, the AECB requires special licenses, which, at this writing (June 1979) BNGS A does not have.
- b) Adjuster Rods

Solid rods of a neutron absorbing material (Cobalt or Stainless Steel). Normally fully inserted in the reactor thus increasing parasitic absorption (decreasing f). Positive reactivity is provided by withdrawing the adjuster rods.

Advantages:

- Provide flux flattening which must be provided by some other method if booster rods are used for xenon override.
- No significant decrease in reactivity worth over normal lifetime.

Disadvantages:

 Presence of adjusters results in a fuel burnup penalty of ~8%. (The adjusters reduce f, therefore, we must increase one of the other factors. Thus n is increased by not allowing the fuel to burn out as much.

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# Long Term Reactivity Control

The method of long term reactivity control presently in use is the addition of soluble poison to the moderator. While solid rods could be used for this purpose, soluble poison systems are cheaper and cause no flux distortions. However, the addition of poison to the moderator does reduce the flux reaching the ion chambers sufficiently to require that the power reading from out of core ion chambers be corrected for the presence of the poison. Boron in the form of boric acid D<sub>3</sub>BO<sub>3</sub> or gadolinium in the form of gadolinium nitrate  $Gd(NO_3)_3$  ·6H<sub>2</sub>O are the poisons presently in use. Gadolinium has the advantage over boron for Xe load simulation because the neutron burn up rate of the neutron absorbing gadolinium isotopes (Gd155 and Gd157) and the Xe build up are sufficiently complementary that little adjustment of the gadolinium concentration by IX control is necessary during start up. The IX columns are, however, used to remove the reactivity build up of low cross section gadolinium absorption products to limit their accumulation in the moderator.

Using boron to simulate Xe load needs a closely monitored operation of the cleanup circuit to obtain the rapid reduction of boron required (3.5 ppm = 28 mk), boron removal being essentially only dependent on the IX removal rate rather than neutron burn up rate. Much more IX column capacity is also needed for B removal than for the Gd system. Gadolinium is not used at Pickering A as there is some concern that it may lead to high deuterium gas levels in the cover gas system due to increased radiolysis of the moderator.

# Equilibrium Fuel Burn Up

On power refueling is used in all of Ontario Hydro's reactors. This essentially keeps the amount of fissile material constant by replacing irradiated fuel with fresh fuel more or less continually. This system of refueling has several distinct advantages:

- 1) No downtime for refueling
- 2) Better average fuel burnup
- 3) Better flux shaping.
- 4) Failed fuel can be removed easily without a shutdown.

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There are of course some disadvantages mainly the high capital cost of the fueling machines and the maintenance which is required for them.

If the fueling machines are unavailable for some reason, there is a limited time the reactor can continue to operate. A Eruce 'A' reactor normally consumes 0.5 mk/day. (That

is the reactivity worth of the fuel diminishes at that rate). If the LZC were at 50% at full power in an equilibrium fuel condition, about 3 mk of excess positive reactivity would be available. That gives approximately 6 days of operation before we must start inserting the boosters (actually undesirable for this purpose) or reduce the operating power (called derating) or shutdown the reactor.

On the other hand, if you overfuel the reactor you may have to derate the reactor due to the high flux in the area of the new fuel, (called regional overpower).

## Shutdown Systems

Early Candu designs had a single shutdown system. As the design of the reactor became more sophisticated, the requirement for extremely high reliability dictated that two independent shutdown system be provided. There are presently three types of shutdown systems in use.

## 1) Moderator Dump

As the moderator level decreases, the physical size of the active portion of the core decreases. As the core gets smaller, leakage increases ( $\Lambda_{f}$  and  $\Lambda_{\downarrow}$  go down).

## Advantages:

- 1) Simple, fail safe with gravity system.
- 2) Absolute shutdown, with the moderator dumped the core cannot be made critical.

## Disadvantages:

- Slow for a large reactor. The initial reactivity insertion rate may not be adequate to protect the reactor from certain types of accidents. Figure 1 shows reactivity vs time for moderator dump at PNGSA. Note that in the first two seconds only -2mk of reactivity has been inserted.
- 2) Time required to pump the moderator back into the calandria is so long (~50 min. at PNGSA) in a larger reactor that a poison out is quite possible.

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# 2) Shutoff Rods

Hollow cylinders of neutron absorbing material (normally stainless steel sheathed cadmium) which can be gravity dropped into the reactor. Their presence greatly increases parasitic absorption thus reducing the thermal utilization (f).

## Advantages:

- Rapid reactivity insertion as required for protection in certain worst case accidents. Figure 2 shows reactivity vs time for PNGSA shutoff rods. Note that in 2 seconds the rods have inserted -22 mk.
- Rapid recovery from a trip is possible. (~3 minutes to withdraw the rods).

## Disadvantages:

- Limited reactivity depth. As presently designed shutoff rods do not provide enough reactivity for a guaranteed long term shutdown.
- Complex system (relative to dump) subject to mechanical failure. Safety analysis normally assumes that the two most reactive rods don't drop on a trip.

## 3) Poison Injection

Poison (Gadolinium) is injected into the moderator under high pressure. This causes a large reduction in the thermal utilization (f).

## Advantages:

- Rapid insertion of reactivity. Figure 3 shows reactivity vs time for BNGSA poison injection system. Note that -33 mk is inserted in 1.5 seconds. Total worth is approximately -675 mk.
   Disadvantages:
- Poison must be removed from the moderator by ion exchange which is costly and slow (~12 hours). If poison injection shuts down the reactor, a Xenon poison out will occur before the moderator poison can be removed.



FIGURE 2



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		NPD	Douglas Pt.	Pickering A	Bruce A	Pickering B	Bruce B
Automatic Reactor Regulation(1)	Automatic Reactor Primary Regulation <sup>(1)</sup>		4 Control Absorbers (3mk)	l4 Liquid Control Zones (5.4mk)	l4 Liquid Control Zones (6mk)	14 Liquid Control Zones (6mk)	14 Liquid Control Zones (6mk)
	Secondary	NONE	Moderator Level : Control	Moderator Level Control	4 Control Absorbers (7mk)	4 Control Absorbers (10mk)	4 Control Absorbers (9.5mk)
Xenon Override		l Booster Rod (2.4mk)	8 Booster Rods (10mk)	l8 Adjuster Rods (18mk)	l6 Booster Rods (15mk)	21 Adjuster Rods (18mk)	24 Adjuster Rods (18mk)
Long Term Reactivity Control	т. (	Moderator Level Control	Moderator Poison Addition (Variable reactivity depending on poison Concentration)				
Equilibrium Fuel Burn up		All Stations use	on power refu	eling			
Shutdown Systems	SDS l	Moderator Dump	Moderator Dump	ll Shutoff Rods (24mk)	30 Shutoff Rods (40mk)	28 Shutoff Rods (48mk)	32 Shutoff Rods (69mk)
	SDS 2	NONE	NONE	Moderator Dump(2)	Poison Injection (55mk in 2.9s)	Poison Injection (N/A)	Poison Injection (55mk in 2.9s)

NOTES: (1) The primary system is normally used for reactor regulation. If the primary system is unavailable or has insufficient reactivity depth, the secondary system will act automatically.

(2) Operation of the dump system at Pickering A is not entirely independent of the shutoff rods.

# TABLE 3

## Reactivity Control Systems

## ASSIGNMENT

- A Bruce reactor trips inserting 40mk due to the shutoff rods. Using the information in Table 2 and assuming an equilibrium fuel condition, would you expect the reactor to remain shutdown (subcritical) if the heat transport system was kept at normal operating temperature? If the heat transport system was cooled down? Justify your answers.
- Both methods of Xenon Override require derating when used. Explain why.
- 3. Simple chemical analysis for boron or gadolinium is not considered sufficient to determine the reactivity worth of moderator poison, explain why.

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THE APPROACH TO CRITICAL

The initial approach to criticality is a procedure undertaken with a great deal of respect because the reactor is in a potentially dangerous condition. The reasons for this are:

- 1. Available reactivity is near its maximum value since there has been no fuel burnup and there are no fission products present. This excess positive reactivity is compensated for by moderator poison; however, the poisons are removable, hence the possibility of a large positive reactivity insertion exists.
- Normal nuclear instruments (ion chambers and/or flux detectors) will be "off scale" at their low end (~10<sup>-5</sup>% of full power); therefore, the regulating system will not automatically control the reactor.
- 3. Although startup instruments (He-3 or  $BF_3$  detectors) will be wired into the shutdown systems, their response becomes increasingly longer as the flux levels decrease.
- 4. The critical value of the control variable is not precisely known. For example if the approach to critical is being made by raising moderator level, the critical level is only a design estimate. (These are generally quite accurate.)

During the approach to criticality the reactor will by definition the subcritical. Therefore, you should review the behaviour of neutron power in a subcritical reactor. (lesson 227.00-9).

## The First Approach to Critical

The most common method in the past has been to raise moderator level until enough fuel was covered to sustain a chain reaction. More precisely,  $k_{\infty}$  was fixed and the leakage was gradually reduced until k was exactly 1. This procedure was used at NPD, Douglas Point and Pickering Units 1 and 2.

Alternatively, with a high enough poison concentration in the moderator to ensure that criticality cannot be pospible. Start at a certain moderator level (nominally near full calandria). (This is known as guaranteed shutdown state.

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The poison is then gradually removed until criticality is reached. In this case, the leakage is nearly constant, and kis increased by raising the value of f, the thermal utilization, until k becomes equal to 1. This was the procedure used at PNGS 'A' Unit 3, and BNGS 'A', which of course doesn't have moderator level control at all. It will be used on all future reactors.

## Pickering Unit 1

The conditions prior to the startup were as follows:

- A boron concentration of 7.25 ppm was chosen for the moderator system to achieve a first critical level just above 4 m. This figure was obtained from design calculations.
- 2. All adjuster rods were fully inserted, and all light water zone compartments were full.
- 3. The heat transport system was cold (46° C) and pressurized with the normal number of heat transport system pumps (12) running.
- 4. Three fission counters (designated NT9, NT8 and NT7), mounted in an aluminum tube, and one He-3 counter were located in channel U-ll which was otherwise empty (ie, no fuel or heat transport fluid).
- 5. Three more He-3 counters were mounted outside the core (in the ion chamber housing) to test a proposal to startup later Pickering units using out-of-core instruments alone.
- 6. The count rates from the in-core neutron counters were determined by feeding their output pulses to scalers, which counted all pulses arriving in a preset time (of the order of 5 minutes at low count rates).
- 7. The protective system trips were set on the output of ratemeters connected to the fission counters NT8 and NT9 and the He-3 counter in channel U-11. Trip levels were always maintained at about <u>one decade</u> above the prevailing count rate.

The approach to critical was monitored by devising an (approximately) linear plot which could readily be extrapolated to predict the critical moderator level. From lesson 227.00-9 recall that:

$$P_{\infty} = \frac{P_0}{1-k} = - \frac{P_0}{\Delta k}$$

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Since the count rate on any detector is proportional to  $P_{\infty}$ , we can now write:

$$\frac{1}{\text{count rate}} \propto 1 - k \quad \alpha \Delta k$$

Since  $\Delta k$  is a direct function of moderator level (as level increase, k increases), we can plot the reciprocal count rate versus moderator level as shown in Fig. 1.



Figure 1

Approach-to-Critical Graph

The intercept of this curve with the moderator level axis should therefore give the critical level.

Pickering A, Unit 3 and all Bruce A units obtained initial criticality by removing poison (boron or a combination of boron and gadolinium) from the moderator. In these cases the moderator was at full tank throughout the startup. The multiplication constant (k) is a direct linear function of poison concentration (1 ppm boron = 8.85 mk; 1 ppm gadolinium = 31.42 mk). Because of this, total poison load may be directly calculated and a plot of poison load versus inverse count rate is a straight line. Figure 2 is a plot of inverse count rates from the incore detectors for the Bruce A, Unit 1 initial criticality. Note that they all give straight lines which accurately predicted the poison concentration at criticality.







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These types of approaches do not have to be repeated for every startup. Once sufficient fission products have been builtup to give a significant photoneutrons source, (ie, actual neutron power  $>10^{-5}$ %) the reactor may be started up using installed instrumentation and automatic regulation.

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FAILED FUEL MONITORING

The operating conditions in CANDU reactors impose severe stresses on the fuel. Sometimes fuel cladding failures occur. Failures vary in size from minute hair-line cracks to large ruptures or holes in the zircaloy cladding. It is important to locate failures as soon as possible. Activity monitoring systems are used (a) to detect the fuel failures and (b) to locate the fuel channel containing the defective element.

## Reasons for Activity Monitoring

Upon fuel failure, some of the active fission products, and uranium and plutonium are able to escape, and go into the primary coolant circuit. There is a limit on the amount of activity allowed to circulate in the coolant, and this is stipulated by the AECB through federal radiation protection regulations. The reasons for being concerned about activity release to the heat transport system are listed below:

- (1) The released activity, which depends upon the defect size and rate of fuel burnup can be extremely high and may constitute a health hazard because (a) increased radiation from the primary coolant circuit equipment may reach unacceptable levels for operating personnel and (b) coolant water leaks and contamination of coolant circuit equipment would bring maintenance staff into direct contact with highly active fission products and plutonium.
- (2) A fuel cladding failure may result in a distorted element making removal from the reactor difficult and also expensive in terms of a clean up operation and a reduced capacity factor.
- (3) Fissile uranium or plutonium released from a fuel failure may become plated out on parts of the coolant circuit. In the presence of a neutron flux they will continue to produce and release fission products into the coolant, making the detection of further fuel failures more difficult.

## Properties of the Fission Products

To detect and locate a failed fuel element the activity of the fission products is measured with a suitable detector situated in the coolant circuit. To determine which fission products we should monitor, the factors to be considered are decay modes, half lives and recirculation behaviour in the coolant, of the most important fission products, and also the background activities present in the coolant.

The fission products may be conveniently classified into three distinct groups according to their chemical properties. These groups are shown in Tables I, II and III which give the corresponding half lives and decay modes of the nuclides.

## Group I Gaseous Fission Products

These gases, comprising various isotopes of Kr and Xe (Table I) that are fairly easily released into the coolant through a defect, recirculate continuously in solution until they decay. The decay mode is by  $\beta$ ,  $\gamma$  emission, and the  $\gamma$  energies are unique for each isotope.

## Gaseous Fission Products (GFP)

Nuclide	Half Life	
<sup>87</sup> Kr	76 min	
<sup>8 8</sup> Kr	2.8 h	<i>P</i> w
<sup>8 5 m</sup> Kr	4.4 h	decay
<sup>1 3 5 m</sup> Xe	15.6 min	modes
<sup>138</sup> Xe	l4 min	
<sup>1 3 5</sup> Xe	9.2 h	
<sup>1 3 3</sup> Xe	5.7 d	
		• • • •

## TABLE I

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# Halogen Fission Products

# TABLE II

	Nuclide	Half Life	
Short Lived Long Lived	$\begin{cases} {}^{\theta} {}^{7} Br \\ {}^{\theta} {}^{8} Br \\ {}^{1} {}^{3} {}^{7} I \\ \\ {}^{1} {}^{3} {}^{2} I \\ {}^{1} {}^{3} {}^{4} I \\ {}^{1} {}^{3} {}^{5} I \\ {}^{1} {}^{3} {}^{3} I \\ {}^{1} {}^{3} {}^{1} I \\ \\ {}^{1} {}^{3} {}^{1} I \\ \end{array}$	56 s 16 s 23 s 2.4 h 52 m 6.7 h 21 h	rs $\beta, \gamma$ decay modes
		0.1 U	ļ

# Depositing Fission Products (DFP)

# TABLE III

Nuclide	Half Life	
	<pre>78 h 67 h Short Lived 66 Days 40 Days 1 Year 33 Days 285 Days</pre>	β,γ Decay Mode

## Group II Halogen Fission Products

This group (Table II) consisting of volatile iodine and Bromine isotopes, is fairly easily released through a defect and subsequently carried in the coolant as negative ions ie, anions. These ions may be removed to some extent in the anion ion exchange (IX) column situated in a clean up line normally in parallel with the primary coolant flow line as shown for the typical activity monitoring system at NPD in Figure 1. The decay mode is by  $\beta,\gamma$  emission but an important feature of the three short lived isotopes listed here is that they are delayed neutron precursors ie, the  $\beta$  decay leads to a highly excited nucleus which is then able to decay further by emitting a delayed neutron.

## Group III Depositing Fission Products

This group of  $\beta,\gamma$  emitting non volatile nuclides (Table III) is not so easily released through a defect as the above groups. Because of their low solubility they plate out (deposit) on the walls of the boiler. They are also removed in the anion ion exchange (IX) column so that most of the activity from this group does not recirculate.

## Fission Product Yield

The relative quantities of the above fission product nuclides produced by fission may be determined by inspection of Figure 2. This double peaked curve shows the % production of the fission products from fission of U-235. The positions of the three groups of fission products are indicated, the yields of those important in activity monitoring are between 1% and 7%.

# Choice of Fission Products for Detection of Fuel Failures

It can be seen that there is a large variation in the half lives of the various fission products and this fact enables us to distinguish between a minor and major fuel cladding failure. A finite time is taken for the fission products to diffuse through a small defect in the cladding during which time the fission products with short half lives will have been decaying. A larger defect occurring under the same steady conditions will release its fission products more quickly and the activity in the coolant will now contain a higher proportion of short lived fission products than in the case of the smaller defect. In order to monitor for detection of a fuel cladding failure under steady reactor operation a fission product with a long life is chosen.

At NPD for example Iodine -133 with a half life of 20hrs is used to monitor all 132 channels in bulk for detection of failures, the set-up used being shown in Figure 1. The iodine monitor is actually called the GFP monitor although this isotope (I-133) is actually in the halogen group of fission products.

At Pickering, with 390 channels, bulk GFP monitors are also used to detect fuel failures in each of the two heat transport loops and utilize various fission products from groups I and II, the most common ones being <sup>131</sup>I, <sup>133</sup>I, <sup>87</sup>Kr and <sup>135</sup>Xe.

# Choice of Fission Products for Location of Failures

Only a small portion of the activity from a fuel defect will be removed by the IX columns for each cycle of the circulating coolant. This means that the activity from a defect will spread throughout all the channels. To locate the defect, we must be able to discriminate between freshly released activity and recirculating activity.

Detection of GFP tells us only that there is a fuel failure and we use detection of the DFP or of the DN groups to home in on the defect.

The depositing fission product (DFP) group, in particular the isotopes <sup>99</sup>Mo and <sup>132</sup>Te, as well as being removed by decay have the property of plating out (depositing) on the boiler tubes and in monel traps used for the efficient deposition of these fission products in the DFP monitors themselves. Hence, the DFP's are more effectively removed from the coolant than the GFP's which are removed only in the clean up line and by natural decay. A DFP monitoring system is usually arranged so that each channel (or zone of channels) may be scanned individually in order to locate the defective channel (or zone). Good discrimination against the circulating activity background can also be obtained by monitoring the delayed neutrons from the precursors <sup>137</sup>I and <sup>87</sup>Br. This good discrimination results because the delayed neutron (DN) monitors are insensitive to  $\gamma$  rays and the short half lives of the precursors give an inherently low recirculating activity background.

The short half lives of the DN precursors are advantageous in discrimination but cause the system to be rather insensitive in the case of small fuel cladding failures. When small defects are suspected the reactor power is cycled, this should have the effect of greatly increasing the proportion of short lived fission products which leak into the coolant. Very small defects can then be detected by the DN monitors.

Monitoring of the delayed neutrons using the reactor power cycling techniques can be used to locate defective elements in individual or groups of channels in conjunction with a DFP monitoring system. Examples of specific systems are briefly described below.

For location purposes at NPD a DFP monitor is used detecting <sup>9</sup><sup>9</sup>Mo and <sup>132</sup>Te, it is able to scan all reactor channels independently so that single defective channels may be found. In addition, a DN monitor making use of <sup>137</sup>I and <sup>97</sup>Br is used to monitor two individual experimental fuel channels.

The locating system at Pickering utilizes 12 DFP monitors, again detecting <sup>9</sup>Mo and <sup>132</sup>Te, to determine the location of a failure to within the zone of channels feeding one of the 12 boilers. When the DFP monitor has indicated the presence of a failure in a particular zone the fuelling machines are then located onto each suspected channel and a search made using a DN monitor on the fuelling machine heads, until the defective channel is found.

# Difficulties Encountered with Fuel Failure Monitoring Systems

The circulating background activity problem results in poor channel discrimination for the GFP system necessitating the use of a DFP and/or DN system for location purposes. Other background activities are also present in the system making the problem of both detection and location of failures more difficult. One of these activities is the manufacturing contamination from uranium dioxide on (or in) the cladding. The contaminant uranium undergoes fission and results in the continual release of fission products into the coolant without the cladding being defective. Additional short lived  $\gamma$  activity is continually present in the coolant, independent of fuel defects, from the presence of <sup>19</sup>O ( $t_2^1 = 29s$ ) and <sup>16</sup>N ( $t_2^1 = 7.3s$ ) produced by neutron activation of the oxygen in the heavy water coolant. Another  $\gamma$  activity source in the coolant, independent of fuel defects, is from activated corrosion products such as <sup>60</sup>Co, <sup>59</sup>Fe, <sup>124</sup>Sb, <sup>65</sup>Zr and <sup>54</sup>Mn resulting from the corrosive effect of coolant on the reactor components. These corrosion products circulate mainly as cations and are removed by a cation ion exchange column situated before the iodine monitor, as shown in Figure 1.

In the case of the DFP and GFP monitors all these additional  $\gamma$  activities present an undesirable spectrum of  $\gamma$  background which makes the electronic discrimination, used to detect a single  $\gamma$  ray energy of a particular isotope, more difficult.

For the DN monitors there is little direct background from these additional  $\gamma$  ray activities but the  $\gamma$  activity of  ${}^{16}$ N in the coolant circuit presents a problem. This isotope emits  $\gamma$ rays of 6.1 and 7.1 MeV which are of sufficient energy to cause emission of photo-neutrons in the D<sub>2</sub>O coolant by the  $\gamma$ (D,p)n reaction. This results in an undesirable neutron background in the DN monitors reducing their sensitivity. 227.00-15

## ASSIGNMENTS

- 1. What may be the operational consequences of neglecting to monitor the fuel channel activity?
- 2. What properties of I<sup>133</sup> make it a useful isotope in a fuel monitoring system?
- 3. What properties of the depositing fission products make them useful in a fuel defect locating system.
- 4. What are the sources of background activities in an activity monitoring system.

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	APPENDIX A
SYMBOL	NAME
β	Delayed Neutron Fraction
γ	Fission Product Yield (normally with subscript to denote particular isotope, eg, $\gamma_{\rm xe})$
ε	Fast Fission Factor
η	Reproduction Factor
$\Lambda_{f}$	Fast Neutron Non-Leakage Probability
Λt	Thermal Neutron Non-Leakage Probability
ν	Neutron Yield per Fission
к,	Mean Log Energy Decrement
Σ	Macroscopic Neutron Cross Section
σ	Microscopic Neutron Cross Section
Τ	Reactor Period
φ	Neutron Flux
f	Thermal Utilization Factor
k	Neutron Multiplication Factor
$\Delta \mathbf{k}$	Reactivity
l	Neutron Lifetime
N	Number Density
S。	Primary Source Strength
${\tt S}_\infty$	Effective Source Strength
р	Resonance Escape Probability

# APPENDIX B

## Properties of the Elements and Certain Molecules

Element or molecule	Symbol	Atomic number	Atomic or molecular weight*	Nominal density, gm/cm <sup>3</sup>	Atoms or molecules per cm <sup>3</sup> †	σ <sub>a</sub> ,‡ barns	σ.,‡ barns	∑a,† cm <sup>-1</sup>	Σ.,† cm <sup>-1</sup>
Actinium	Ac	89	227	·		800			
Aluminum	AI	13	26.9815	2.699	0.06024	0.235	1.4	0.01416	0.08434
Antimony	Sb	51	121.75	6.62	0.03275	5.5	4.3	0.1801	0.1408
Argon	Ar	18	39.948	Gas		0.63	1.5		
Arsenic	As	33	74.9216	5.73	0.04606	4.5	6	0.2073	0.2764
Barium	Ba	56	137.34	3.5	0.01535	1.2	8	0.01842	0.1228
Beryllium	Be	4	9.0122	1.85	0.1236	0.0095	7.0	0.001174	0.8652
Beryllium oxide	BeO		25.0116	2.96	0.07127	0.0095	6.8	0.0006771	0.4846
Bismuth	Bi	83	208.980	9.80	0.02824	0.034	9	0.0009602	0.2542
Boron	В	5	10.811	2.3	0.1281	759	4	97.23	0.5124
Bromine	Br	35	79.909	3.12	0.02351	6.7	6	0.1575	0.1411
Cadmium	Cd	48	112.40	8.65	0.04635	2450	7	113.6	0.3245
Calcium Carbon	Ca	20	40.08	1.55	0.02329	0.43	3.0	0.01002	0.06987
(graphite)**	C	6	12.01115	1.60	0.08023	0.0034	4.8	0.0002728	0.3851
Cerium	Ce	58	140.12	6.78	0.02914	0.7	9	0.02040	0.2623
Cesium	Cs	55	132.905	1.9	0.008610	30	20	0.2583	0.1722
Chlorine	Cl	17	35.453	Gas		33	16		
Chromium	Cr	24	51.996	7.19	0.08328	3.1	3	0.2582	0.2498
Cobalt Columbium	Co	27	58.9332	8.8	0.08993	37	7	3.327	0.6295
(see monum)	C	20	67.64	8.04	0.08402	20		0.000	
Deuterum		1	201410	6.90 Gao	0.06493	3.8	1.2	0.3227	0.6115
Duarouum		66	162.50	Qas 8 sc	0.03173	0.0003	100	20.92	
Erburn	Dy E-	00	102.50	0.30	0.03172	940	100	29.82	3.172
Euronum	EI E.	60	167.20	9.16	0.03203	160	15	5.125	0.4805
Eluorine	r r	05	121.90	5.22	0.02069	4300	8	88.97	0.1655
Gadolinium	Gđ	64	157.25	Gas 7.05	0.01046	0.0098	3.9		
Gallum	Ga	21	137.25	7.95	0.03045	46,000	4	1401	0.1218
Germanium	Ge	32	77 59	5.76	0.03105	3.0	,	0.1532	0.1334
Gold	Au	70	196.967	10 77	0.04447	4.4	3	0.1067	0.1334
Hafnium	Hſ	72	178 49	13.32	0.03507	90.0	9.3	5.830	0.5494
Heavy water t	D.0		20.0276	1 105	0.04308	0.0010	12.6	4.733	0.3000
Helium	He	2	4 0076	6.105 6.25	0.03525	< 0.0010	13.0	5.525 X 10 V	0.4519
Holmium	Но	67	164 930	8 76	0 03100	<u>65</u>	0.8	2 070	
Hydrogen	н	I	1.008665	Gas	0.03133	0.322		2.079	
Illinium (see promethium		•	1.000000	Uas		0.332			
Indium	In	49	114.82	731	0.03834	104		7 470	0.00436
Iodine	I	53	126 9044	4.03	0.03834	64	2.2	7.438	0.08435
Iridium	Ir	77	192.2	22.5	0.02040	460	5.0	0.1498	0.08242
Iron	Fe	26	55 847	7.87	0.08487	2 400	11	32.43	0.0226
Krypton	Kr	36	83.80	Gas	0.08407	2.55	11 77	0.2147	0.9330
Lanthanum	La	57	138.91	619	0.02684	24 90	1.4	() 3340	11 411 30
Lead	Ph	82	203 973	11.34	0.02054	0.7	13	0.004(02	0.4026
Lithium	Li	3	6 9 3 9	0.53	0.03548	71	11	0.005692	0.3683
Lutetium	Lu	71	174.91	9.74	0.03354	80	1.4	3.200	0.0644
Magnesium	Mg	12	24.312	1.74	0.04310	0.063	4	4.083	0 1734
Manganese	Mn	25	54.9380	7.43	0.08145	13.3	22	1 022	0.1/24
Mercury	Hg	80	200.59	13.55	0.04068	360	20	1.005	0.10/3
Molybdenum	Mo	42	95.94	10.2	0.06403	2.6	7	0 1665	0.0130
Neodymium	Nd	60	144.24	6.98	0.02914	50	16	1 457	0.4663
Neon	Ne	10	20.183	Gas		0.032	24		0.4002
Nickel	Ni	28	58.71	8.90	0.09130	4.6	17.5	0.4200	1 507
Niobium	Nb	41	92.906	8.57	0.05555	1.1	5	0.06111	0.2778
Nitrogen	N	7	14.0067	Gas		1.85	10		0.2770

- 1 -

Element or molecule	Symbol	Atomic number	Atomic or molecular weight*	Nominal density, gm/cm <sup>3</sup>	Atoms or molecules per cm <sup>3</sup> †	σa,‡ barns	σ.,‡ barns	$\Sigma_{a}$ ,† cm <sup>-1</sup>	Σ,,† cm -1
Osmium	Os	76	190.2	22.5	0.07124	15	11	1.069	0.7836
Oxygen	0	8	15.9994	Gas		< 0.0002	4.2		
Palladium	Pd	46	106.4	12.0	0.06792	8	3.6	0.5434	0.2445
Phosphorus									
(yellow)	P	15	30.9738	1.82	0.03539	0.19	5	0.006724	0.1770
Platinum	Pt	78	195.09	21.45	0.06622	10	10	0.6622	0.6622
Plutonium	Pu	94	239	19.6	0.04939	$\sigma_a = 1015$ $\sigma_c = 741$	9.6	49.88 36.55	0.4741
Polonium	Po	84	210	9.51	0.02727	•,			
Potassium	K	19	39,102	0.86	0.01325	2.1	1.5	0.02783	0.01988
Praseodymium	Pr	59	140.907	6.78	0.02898	12 -	4	0.1965	0.1159
Promethium	Pm	61							
Protactinium	Pa	91	231			210			
Radium	Ra	88	226	5.0	0.01332	20		0.2664	Ì
Rhenium	Re	75	186.2	20	0.06596	85	14	5.607	0.9234
Rhodium	Rh	45	102.905	12.41	0.07263	155	5	11.26	0.3632
Rubidium	Rb	37	85.47	1.53	0.01078	0.73	12	0.007869	0.1294
Ruthenium	Ru	44	101.07	12.2	0.07270	2.5	6	0.1818	0.4362
Samarium	Sm	62	150.35	6.93	0.02776	5800	5	161.0	0.1388
Scandium	Sc	21	44.956	2.5	0.03349	23	24	0.7703	0.8038
Selenium	Se	34	78.96	4.81	0.03669	12	11	0.4403	0.4036
Silicon	Si	14	28.086	2.33	0.04996	0.16	1.7	0.1164	0.08493
Silver	Ag	47	107.870	10.49	0.05857	63	6	3.690	0.3514
Sodium	Na	11	22.9898	0.97	0.02541	0.53	4	0.01347	0.1016
Strontium	Sr	38	87.62	2.6	0.01787	1.3	10	0.02323	0.1787
Sulfur									
(yellow)	S	16	32.064	2.07	0.03888	0.52	1.1	0.2022	0.04277
Lantalum	Ta	73	180.948	16.6	0.05525	21	5	1.160	0.2763
Technetium	Tc	43	99			22			
Tellurium	Te	52	127.60	6.24	0.02945	4.7	5	0.1384	0.1473
Terhium	Tb	65	158.924	8.33	0.03157	46		1.452	
Thallium	T1	81	204.37	11.85	0.03492	3.3	14	0.1152	0.4889
Thorium	Th	90	232.038	11.71	0.03039	7.4	12.6	0.2249	0.3829
Thulium	Tm	69	168.934	9.35	0.03314	125	7	4.143	0.2320
Tin	Sn	50	118.69	7.298	0.03703	0.63	4	0.02333	0.1481
Titanium	Ti	22	47.90	4.51	0.05670	6.1	4	0.3459	0.2268
Tungsten	W	74	183.85	19.2	0.06289	19	1 5	1.195	0.3145
Uranium	U	92	238.03	19.1	0.04833	$\sigma_{11} = 7.6$	8.3	0.3673	0,4011
				-		$\sigma_f = 4.2$		0.2030	
Vanadium	V	23	50.942	6.1	0.07212	4.9	5	0.3534	0.3606
Water	$H_2O$		18.0167	1.0	0.03343	0.664	103	0.02220	3.443
Xenon	Xe	54	131.30	Gas		24	4.3		
Ytterbium	Yb	70	173.04	7.01	0.02440	37	12	0.9208	0.2928
Yttrium	Y	39	88.905	5.51	0.03733	1.3	3	0.04853	0.1120
Zinc	Zn	30	65.37	7.133	0.06572	1.10	3.6	0.07229	0.2366
Zirconium	Zr	40	91.22	6.5	0.04291	0.18	8	0.007724	0.3433

• Based on  $C^{12} = 12.00000$  amu.

\* Based on  $C^{12} = 12,0000$  amu. † Four-digit accuracy for computational purposes only; last digit(s) usually is not meaningful. ‡ Cross sections at 0.0253 eV or 2200 m/sec. The scattering cross sections, except for those of H<sub>2</sub>O and D<sub>2</sub>O, are measured values in a thermal neutron spectrum and are assumed to be 0.0253 eV values because  $\sigma_i$  is usually constant at thermal energies. The errors in  $\sigma_i$  tend to be large, and the tabulated values of  $\sigma_i$  should be used with caution. (From BNL-325, 2nd ed., 1958 plus supplements 1 and 2, 1960, 1964, and 1965.) \*• The value of  $\sigma_a$  given in the table is for pure graphite. Commercial reactor-grade graphite contains varying amounts of contaminants and  $\sigma_a$  is somewhat larger, say, about 0.0048 barns, so that  $\Sigma_a \approx 0.0003851 \text{ cm}^{-1}$ . †† The value of  $\sigma_a$  given in the table is for pure D<sub>2</sub>O. Commercially available heavy water contains small amounts of ordinary water and  $\sigma_a$  in this case is somewhat larger.

case is somewhat larger.

APPENDIX C

# NUCLIDES AND ISOTOPES

ACCOMPANYING THE CHART OF THE NUCLIDES REVISED 1969 BY DR. NORMAN E. HOLDEN AND F. WILLIAM WALKER KNOLLS ATOMIC POWER LABORATORY, SCHENECTADY, N.Y.

# INTRODUCTION

The earliest discussion of the atomic hypothesis is attributed to the ancient Greek philosophers who speculated about the mysteries of nature. In the fifth century B.C., Democritus believed that elementary substances (earth, water, fire, and air) were formed by minute individual particles called atoms. This vague philosophical speculation was given reality when John Dalton, between 1803 and 1808, showed how to determine the weights of different atoms relative to one another.

In 1816, William Prout believed (based on the few atomic weights known) that all atomic weights were whole numbers and integral multiples of the atomic weight of hydrogen. He thought that all elements might be built up from hydrogen. His concept lost favor when elements such as chlorine were definitely shown to have noninteger atomic weights.

#### PERIODIC PROPERTIES OF ELEMENTS

In 1869, Dmitri Mendeleev published a short note on the periodic regularity of chemical elements. He arranged the elements in rows according to the magnitude of their atomic weights, beginning with the smallest weight. Elements that appeared in the same vertical column showed a remarkable similarity in their chemical properties. Mendeleev hypothesized that deviations from the expected periodicity were due to chemists' failure to discover some elements in nature. He predicted the properties of gallium, scandium, and germanium, which were subsequently discovered. Pairs of elements (for example, tickel and cobalt) that did not fit the periodic properties of their columns were interchanged so that they would correspond. He argued that the atomic weight measurements for these elements must be in error. It is now known that the atomic number (see page 2), rather than the atomic weight, is the correct basis for the periodicity in the chemical properties of the elements. By coincidence, the list of elements ordered by atomic weight usually agrees with the list ordered by atomic number, except for the few cases observed by Mendeleev.

## **NEW PHENOMENA**

Toward the end of the nineteenth century, the successes in chemistry, together with those of classical mechanics and electromagnetic theory, convinced some individuals that classical physics was a "closed book" and that workers in the field would henceforth merely advance existing knowledge to the next decimal place. This attitude changed in 1895 when Wilhelm Roentgen discovered X-rays and in 1896 when A. Henri Becquerel discovered natural radioactivity. Since such phenomena could not be explained by existing theories of matter, they created great interest.

In 1902, Ernest Rutherford and Frederick Soddy, in their theory of radioactive disintegration, proposed that radioactivity involves changes occurring within the atom. Their view met strong opposition because it was considered contrary to the established view on the permanency of the atom.

### EARLY MODELS OF ATOMIC STRUCTURE

Early experiments in the investigation of atomic structure disclosed three different types of radioactivity. called alpha, beta, and gamma radiation. Alpha rays were found to be positively charged helium ions; beta rays were found to be negatively charged electrons; and gamma rays were highenergy electromagnetic waves. In a magnetic field, the alpha rays were deflected in one direction, the beta rays deflected in the opposite direction, and the gamma rays not deflected at all.

The discovery of radioactivity and Sir Joseph Thomson's proof of the independent existence of the electron were the starting points for theories of atomic structure. Thomson proposed one of the first models of the atom. His "plum pudding" model of internal structure depicted the atom as a homogeneous sphere of positive electric fluid (the pudding) in which were imbedded the negatively charged electrons (the plums). In this model the negatively charged electrons, which repel each other and which are attracted to the positive charge, assume certain stable positions inside the atom. If the electron distribution is disturbed by an external force, e.g., the violent collisions between atoms in a hot gas, the electrons vibrate about their equilibrium positions and emit electromagnetic radiation.

The homogeneous-atom concept was proved incorrect when Rutherford performed a series of experiments with a beam of high-speed alpha particles fired at a very thin metal foil. Most of the alpha particles passed straight through the foil or were scattered or deflected only slightly from their original paths. A small percentage of alpha particles were significantly deflected, however, with some alphas reversing their directions.

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The Thomson model, in which the positive charge was uniformly distributed throughout the atom, would never permit a sufficiently large concentration of this charge in one region to affect the alpha particles significantly. Rutherford thought that "it (the experimental result) was about as credible as if you had fired a 15-inch shell at a piece of tissue paper and it came back and hit you."

To explain these results, Rutherford postulated that the atom does not consist of a uniform sphere of positive electrification, but that the positive charge is concentrated in a small region called the nucleus, at the center of the atom. In his dynamic planetary model, the nucleus plays the role of the sun and the electrons correspond to individual planets of the solar system revolving about the sun. This model, along with the classical physical laws of electricity and mechanics, provided an adequate explanation of the alpha particle's scattering. Subsequent experiments performed on seven different scattering materials and at different alpha energies verified Rutherford's theory.

Electromagnetic theory demands that an oscillating or revolving electric charge emit electromagnetic waves. Such emission results in the loss of energy by the emitting particle. Applied to Rutherford's electrons, this energy loss would cause a steady contraction of the system since the electrons would spiral into the central nucleus as their rotational energy was dissipated. This process would occur very rapidly and would directly contradict the permanent existence of atoms. Also, if the radiation pattern produced by the atom were related to the energy radiated by its moving electron, this radiant energy would be changing with the radius of curvature of the electron's path. The pattern would consist of a continuous range of wavelengths instead of the well-defined discrete wavelengths that are characteristic of each element.

## BOHR ATOM

Since the known stability of atomic systems could not be reconciled with classical principles of mechanics and electrodynamics, Niels Bohr in 1913 reasoned that classical physics laws must be wrong when applied to the motion of the electron in the atom. Max Planck revealed an essential limitation in the theories of classical physics in 1901 when he introduced the concept of discrete amounts of energy (the energy quantum) in his quantum theory of heat radiation. Albert Einstein had applied this concept to light in 1905, when he described the photoelectric effect. The quantum theory states that electromagnetic radiation (of which light is one form) must be emitted or absorbed in integral multiples of these energy quanta. Bohr coupled Rutherford's atom with the quantum theory to produce his quantum theory of atomic structure.

Since a body that spins about its own axis or revolves in an orbit about a central point possesses angular momentum. Bohr assumed that the electron's angular momentum was restricted to certain values (he quantized the angular momentum). Each of the restricted values, which was described by a principal quantum number, n. would specify a particular circular orbit. An atomic system, whose electrons were in given orbits, would not emit electromagnetic radiation even though the particles were accelerating. The whole atom was said to be in a stationary state. Such an assumption is contrary to classical electrodynamics as mentioned earlier. Electromagnetic radiation would be emitted or absorbed only when an electron changed from one allowed orbit to another allowed orbit. The energy difference between the two states would be emitted or absorbed in the form of a single quantum of radiant energy, producing a radiation pattern of a definite frequency  $\gamma$ , related to the energy E by the relation E = h $\gamma$  already postulated by Planck and Einstein.

#### QUANTUM NUMBERS

The quantum theory was further refined in 1916 when Arnold Sommerfeld introduced an azimuthal quantum number, l, where l < n - 1, which permitted discrete elliptical orbits for electrons, in addition to the circular orbits. This change permitted the Bohr model to account for detailed structure in the pattern of radiation emitted by hydrogen and other atoms. To account for the change in the emitted radiation pattern when an atom is exposed to a magnetic field, a magnetic quantum number m (with permitted integral values from -l to -l was added. This quantum number designates different projections of the possible circular or elliptical orbits along the magnetic field direction in space. Finally, a spin quantum number for the electron was postulated by Samuel Goudsmit and George Uhlenbeck to account for the close grouping of two or more spectral lines. An electron was considered to have an angular momentum about its own axis; in mechanical terms, this motion can be thought of as spin. In a magnetic field, the spin axis can have two directions relative to the field.

The orbits in which the electrons move can be described by specifying a set of these four quantum numbers. All electrons with principal quantum 1 are in the innermost orbit, called the number n – K shell. All electrons with n = 2 fall into a second group, called the L shell. The total number of electrons in a shell is limited by the various possible combinations of the other three quantum numbers. When an electron shell is filled, the atom is in a stable configuration (the noble gas configuration) and does not easily undergo chemical reactions. If only one or two electrons are in the last unfilled shell, it is relatively easy for the atom to lose these electrons to another atom whose last unfilled shell has one or two vacancies. The first of these two atoms becomes positively charged (because of the loss of electrons); the second becomes negatively charged (because of the gain of electrons). These atoms can now attract each other and form a compound (ionic bonding).

The periodicity or repetitive structure of the Mendeleev chart is now understood to be due to the number of electrons in the atom. In a neutral atom the number of electrons is balanced by the equal number of protons (hydrogen nuclei with a positive charge and a mass of about 1836 electron masses) in the nucleus of the atom. Note that the atomic number of an element is equal to the number of unit positive charges carried by the nucleus and is not the same as the atomic weight. In 1913, Henry G. J. Moseley determined the magnitude of the nuclear charge by comparing the characteristic X-ray wavelengths of elements. Identification of the atomic number of an element from its highfrequency spectrum provided a rule for fitting newly discovered elements into vacant places on the Mendeleev chart.

In 1923, Louis DeBroglie postulated that, in analogy with light having both a wave and a particle nature, matter should have a wave as well as a particle nature. The wavelength that he predicted for a particle was inversely proportional to the particles' momentum. Clinton J. Davisson and Lester H. Germer experimented with the scattering of electrons from a crystal. They showed that electrons definitely had wave properties with a wavelength corresponding to the value predicted by DeBroglie.

The mechanical picture offered for the classification of stationary states of atoms by the Bohr theory, and its subsequent modification, was handicapped by its reliance on many *ad hoc* postulates and by an inability to explain the intensities of radiation patterns emitted by atoms. A new departure was provided in 1926 by Erwin Schrödinger's establishment of wave-mechanics,\* in which stationary states are conceived as proper solutions of a fundamental wave equation. In advanced theories, the mechanical models are no longer used.

#### ISOTOPES

Experimental investigations in nuclear physics began to require specialized instruments. One of the first of these instruments was the mass spectrograph developed by Francis W. Aston to measure the relative mass of the atoms of an element. This device directed positive ions of an ionized (electrically charged) gas at a photographic plate. The ions were deflected by electric and magnetic fields, working at right angles, so that all particles having the same mass were brought to a focus at a fine line. Heavier ions, having more inertia, were deflected less that were the lighter ions.

With the use of the mass spectrograph, it was discovered that some chemical elements have two or more components, each with its own mass. Natural chlorine, whose atomic weight is fractional (about 35.5), produced two lines on the photographic plate corresponding to masses very close to 35 and 37. No particle was found with a fractional mass (within the experimental error). Components of the same hemical element with different mass numbers are called isotopes. Most elements in their natural state consist of two or more isotopes, although 20 elements have only one isotope: for example, aluminum, cobalt, and gold. Modifying Prout's hypothesis, Aston proposed the whole-number rule which states that all atomic masses are close to integers and that fractional atomic weights are due to the presence of two or more isotopes, each of which has an approximately integral value. On the carbon-12 scale now used, where the atomic weight of carbon-12 is exactly 12 units, all other isotopes have atomic weights close to integers.

With the problem of fractional atomic weights solved, physicists at first believed that nuclei consisted of electrons and protons. A nucleus with an atomic number Z and an atomic mass A would consist of A protons, to account for the total mass, and A minus Z electrons to balance the excess positive charge of the protons. This view of the structure of the nucleus was altered in 1932 when James Chadwick discovered the neutron. This particle has no electric charge and has approximately the same mass as the proton.

It is now believed that neutral atoms consist of N neutrons, Z protons, and Z orbital electrons, with N + Z. Isotopes are nuclides with the same Α : Z but different N. For example, natural hydrogen consists almost entirely of atoms that contain one proton and one electron. However, a small amount (about 0.015 percent) of deuterium (heavy hydrogen) is present in nature; deuterium consists of one proton, one neutron, and one electron. In general, the situation becomes more complex as the heavier elements are encountered. Natural tin, which has atomic number 50, consists of 10 isotopes of masses 112, 114, 115, 116, 117, 118, 119, 120, 122, and 124. These isotopes differ from one another because, although each has 50 protons and 50 electrons, each contains a different number of neutrons (ranging from 62 to 74).

The nucleus is held together by attractive forces between the neutrons and protons. These attractive forces are not completely understood, but it is known that they must be strong enough to overcome the electrostatic repulsion between the protons. Because of this repulsion, however, the ratio of neutrons to protons increases for stable isotopes as the atomic number increases. Among light elements in nature, there is approximately one proton for every neutron. Among heavy stable isotopes, for every two protons there are approximately three neutrons.

As previously mentioned, Aston found that the atomic masses were approximately integers. More accurate measurements indicate that the total mass of a nucleus is always less than the sum of the proton and neutron masses of which the nucleus is composed. In 1905, Einstein had shown that mass, m. was another form of energy, E, expressed by his relationship  $\mathbf{E} = -\mathbf{mc}^2$ , where c is the velocity of light. The mass deficiency of the nucleus is expressed as the nuclear binding energy. The binding energy represents the amount of energy required to break the nucleus into its constituent nucleons. The ratio of the binding energy to the number of particles in the nucleus varies among the stable elements. It is greater for elements with mass numbers between 30 and 120 than it is for very light or very heavy stable elements.

#### ARTIFICIAL RADIOACTIVITY

In 1919, Rutherford's discovery of artificial radioactivity achieved the feat vainly sought by the ancient alchemists, that is, changing one element into another. Rutherford bombarded nitrogen gas with a stream of alpha particles. Some of the alpha particles were absorbed by the nitrogen, protons were emitted, and a different element, oxygen, was formed. The physicist uses symbolic language

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<sup>\*</sup> Wave-mechanics is equivalent to the matrix mechanics developed by Werner Heisenberg in 1975.

to represent the transformation as follows:

$$\frac{14}{7} \xrightarrow{4}{2} \frac{17}{8} \xrightarrow{17}{} \xrightarrow{17}{} H.$$

The superscripts denote the total number of nucleons (number of protons plus neutrons), and the subscripts denote the atomic number (number of protons) in each element. Note that the superscripts on one side of the arrow balance those on the other side. The same is true for the subscripts. The balance represents the conservation of the number of protons and neutrons separately.

This initial discovery has been followed by the construction of large machines designed to accelerate charged particles such as protons and alpha particles to higher energies so that they may be used to bombard nuclei. Among these machines are the Van de Graaff generator, the cyclotron, the betatron, the linear accelerator (linac), and others. Beams of high-energy neutrons can also be produced. Since the neutron is electrically neutral, however, there is no electrostatic repulsion between bombarding neutrons and the positively charged target nuclei. Even thermal neutrons could be used for nuclear reaction studies (thermal neutrons have energies that correspond to the most probable energy for a group of neutrons at  $68^{\circ}$ F, that is, energies in the neighborhood of 0.025 eV).

## **NEUTRON FISSION**

During the investigation of neutron-produced reactions in various target elements, Enrico Fermi and his associates discovered different beta activities (distinguished by half-life) when uranium was used as a target. They assumed that a transuranium element had been produced (that is, an element whose atomic number was greater than 92). In 1938, Otto Hahn and Fritz Strassman, repeating the experiments, discovered part of the activity to be due to barium (atomic number 56). Lise Meitner and Otto Frisch suggested that the uranium nucleus had split into two roughly equal parts, barium and krypton (the latter, atomic number 36), when the uranium captured the incident neutron. This reaction Frisch termed "fission," after the term used to describe the division of cells in a living organism. Since the mass defect (or binding energy) per particle is greater for the residual nuclei, barium and krypton, than for the uranium, neutron fission is accompanied by a large energy release.

For nuclear reactions other than fission, Fig. 1 illustrates the many combinations of incident (or bombarding) and emitted particles, and how each combination changes the original nucleus. This figure is copied from the lower right corner of the chart. A special type of shorthand is used on this diagram to identify the data represented. An example is (p,n) which denotes a reaction in which the nucleus absorbs a proton and emits a neutron. The symbols used are:

n neutron

t triton (hydrogen-3 nucleus)

d deuteron p proton

 $\alpha$  alpha particle

<sup>3</sup>He helium-3 nucleus  $\gamma$  gamma ray

Using these reactions, nuclear physicists have produced far more artificially radioactive isotopes than the stable or radioactive isotopes that occur in nature. The term "nuclide" was proposed by Truman P. Kohman for a species of atom characterized by the number of neutrons and protons that the atom contains. The term is used in this booklet in this general sense to encompass both stable and radioactive species. At present, there are 1675 nuclides known, of which 264 are stable forms of the natural elements. In addition, 65 of the unstable nuclides are found in nature, mainly among the heaviest elements. Active nuclear research, which is conducted in many laboratories throughout the world, causes additions and changes in the list of nuclides. Since the last edition of the chart was published (1966), 8 nuclides, which had been misassigned, have been removed, and 180 new nuclides have been added.

# CHART OF THE NUCLIDES

(Data revised to December 1968; occasional data to June 1969)

The general arrangement of the Chart is similar to that suggested by Emilio Segré and followed in previous editions. Because of its size, the Chart is presented in three overlapping sections. The numbers along the left-hand side, marking the horizontal rows, represent the atomic number Z(the number of protons in each nucleus of that row). Each horizontal row represents one element; the filled spaces indicate the known isotopes of that element. The numbers at the bottom of the vertical columns represent the number of neutrons in each nucleus of that column; the number is designated by N.

Heavy lines on the Chart occur for Z or N equal to 2, 8, 20, 28, 50, 82, and 126. These are the socalled "magic numbers", i.e., the numbers of neutrons (protons) present when a neutron (proton) shell is closed. In analogy with the electron shell model of the atom, a nuclear shell model has been developed for the neutrons and protons within a nucleus. Filled shells represent the most stable configurations. Nuclides having either a closed neutron shell, or a closed proton shell, or both, are most stable.

Spaces shaded in gray represent isotopes that occur in nature and that are generally considered stable. A black rectangular area at the top of a

a, 3n	a, 2n 3 Hei,n	a, n	
p,n	Ρ,γ d,n <sup>3</sup> He,np	a , np 1 , n <sup>3</sup> He ,p	
γ.n n.2n	Original Nucleus	d,p Π,γ 1,np	1, p
γ.ηρ	У , Р	n , p	
n a	n, <sup>3</sup> He		

Fig. 1. Changes Produced by Various Nuclear Reactions

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white square indicates a radioactive isotope that is found in nature. Examples of such isotopes are (1) an unstable nuclide having a lifetime sufficiently long to have prevented the loss by disintegration of all atoms of that particular nuclide that were available at the time the elements were formed, and (2) a short-lived nuclide that is a disintegration product of such a long-lived nuclide. Occasionally one nuclide has both the gray shading and the black top. This indicates an isotope found in nature, such as rubidium-87, that is radioactive with a very long half-life. Squares with smaller black rectangular areas near the top represent members of one of the naturally radioactive decay chains (see page 8). The old symbolic name is inserted in this smaller black area. White squares represent artificially produced radioactive nuclides.

The heavily bordered space at the left side of each horizontal row gives properties of the element as found in nature, including the chemical atomic weight (on a mass scale where the neutral atom of carbon-12  $\dots$  12.00000) and the thermal neutron absorption cross section (see page 6).

Each of the other occupied spaces carries the chemical symbol (a list of these symbols is given on page 7 along with the atomic weights) and the mass number of the nuclide indicated. The mass number, designated by  $A_i$  is the sum of the number of neutrons and protons in the nucleus. The number of neutrons N is equal to the difference between the mass number and the atomic number, that is, A minus Z.

#### STABLE NUCLIDES

Classifying the 264 stable nuclides by the evenor oddness of Z and N gives four possible categories. The first category contains an even number of protons and an even number of neutrons (so called even-even nuclei). The other categories are even-odd, odd-even, and odd-odd. Table 1 shows the number of stable nuclides that fall in each category.

А	Table I.	Distribution	of Stable Nuclides
	Z	N	Number of Stable Nuclides
Even	Even	Even	157
Odd	Even	Odd	53
Odd	Odd	Even	50
Even	Odd	Odd	4
			264

Table 1 shows that for the odd A nuclides there are approximately as many nuclides with an even number of protons (even Z) as with an even number of neutrons (even N). This is evidence that the nuclear force between two nucleons is independent of whether the nucleons are protons or neutrons. Odd-odd stable nuclides are scarce, and they are found only among the lightest nuclides. Their scarcity is due to a "pairing energy" between particles in the same shell. The condition of being in the same shell increases the binding energy of these particles, making them more stable than particles in different shells. An odd-odd nuclide contains at least one unpaired proton and one unpaired neutron which are usually in different shells and hence contribute weakly to the binding. For the lightest nuclei, however, the unpaired neutron and proton are in the same shell.

Diagonals running from upper left to lower right connect nuclides of different elements, which have the same mass numbers. For example, one line could connect calcium-40, which has 20 protons and 20 neutrons, with argon-40, which has 18 protons and 22 neutrons. Nuclides of the same mass number are called isobars; nuclides with the same number of neutrons are called isotones.

#### DATA DISPLAY

The manner of displaying data is explained in the lower right corner of the chart. For stable nuclides, the first line contains the chemical symbol and mass number; the second line presents the atom percent of the natural element that this isotope represents (known as the absolute isotopic abundance); the third line contains the thermal neutron cross section (see page 6); and the fourth line presents the isotopic mass of the neutral atom (the mass of the nucleus and its surrounding electrons). This mass is given in atomic mass units where carbon-12 is assigned a mass of 12.00000.

For long-lived, naturally occurring radioactive nuclides, the first line contains the chemical symbol and mass number, the second line presents the absolute isotopic abundance, and the third line contains the half-life. The half-life is the period of time in which half of the nuclei initially present in a given sample disintegrate. Additional lines present the decay modes (or types) and energies of decay, and the isotopic mass of the nuclide. Energies are given in millions of electron volts (MeV). When more than one mode of decay occurs, the most prominent mode appears first (above, or to the left of, the other modes). When gamma radiation is emitted in more than one decay mode, or if several gamma rays are emitted in one mode, the gamma rays are separated and presented below (or to the right of) their associated decay mode(s).

For radioactive nuclides that are not of the longlived, naturally occurring type, the same information is presented except that the isotopic abundance is omitted and the last line of the pertinent square contains the beta-decay energy instead of the isotopic mass. For the heavy elements, where the major mode of decay is alpha-particle emission, the isotopic mass is retained in the last line. In many squares, a small black triangle appears in the lower right corner to indicate that the nuclide has been formed as a product in the thermalneutron fission of uranium-235.

## METASTABLE STATES

Note that certain squares are divided, for example, the square for aluminum-26. Such divisions occur when a nuclide has one or more isomeric states, that is, when a nuclide has the same mass number and atomic number, but possesses different radioactive properties in different long-lived energy states. On the chart, a long-lived state is arbitrarily defined as a state whose half-life is one microsecond (one-millionth of a second) or longer. The lower energy state is generally referred to as the ground state, the higher state as the isomeric state. Frequently, the ground state is a stable nuclide. If one metastable state exists, it is shown on the left. If two exist, the higher energy state is shown on the

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left, the lower below it or to the right of it, and the ground state to the right of both.

A mode of decay and the decay energy shown in parentheses indicate that the decay results from a short-lived daughter that accompanies its parent. (In a radioactive decay, the original nuclide is called the parent or precursor: the resultant nuclide is called the daughter.) For example, nitrogen-17, with a half-life of 4.14 seconds, decays by negative beta emission (symbol  $\beta$ ) into an exceedingly short-lived state of oxygen-17, which in turn emits a neutron. Thus nitrogen-17 emits "delayed neutrons" with a half-life of 4.14 seconds.

Another example is 17 day palladium-103, which decays by K-electron capture mainly to the 57minute rhodium-103 and, statistically less often, to stable rhodium-103. (K-electron capture occurs when the nucleus captures an electron from the K shell; the symbol is i.) The 57-minute rhodium emits a gamma ray or an internal-conversion electron that corresponds to an isomeric transition of 0.040 MeV. (An internal-conversion process involves the direct transfer of energy from the nucleus to one of the orbital electrons, and the electron is ejected from the atom: the symbol used is e.) On the chart, the delayed gamma ray is assigned to the parent; inclusion of the energy in parentheses indicates that the gamma ray comes from the daughter, but continues to last as long as the disintegrating parent is still present.

A further example is provided by a standard laboratory radionuclide, 30.2-year cesium-137. This long-lived parent decays directly to a short-lived daughter, 2.551-minute barium-137, by negative beta emission. The 6.616-MeV gamma ray which is emitted by the barium is included in parentheses on the cesium square.

# THERMAL NEUTRON CROSS SECTIONS

The Greek letter  $\sigma$  with various subscripts is used to identify the thermal neutron cross sections. The neutron cross section measures the probability of interaction of a neutron with matter. The cross section can be most easily visualized as a crosssectional target area presented to the neutron by the nucleus. The cross section depends upon the type of interaction involved and the energy of the neutron. At thermal energies, a number of reaction types are possible. The thermal neutron absorption cross section (symbol  $\sigma_n$ ) is the sum of the cross sections for all reactions except scattering of the neutron. Cross sections are usually measured in units of barns per atom. A barn is the area of a square a millionth of a millionth of a centimeter on each side ( $10^{-24}$  square centimeters). The most probable reaction (that is, the reaction with the largest cross section) is generally the neutron capture reaction (symbol a.) in which the absorption of the neutron by the nucleus is accompanied by high-energy gamma-ray emission. Occasionally, a proton or an alpha particle may be emitted, or the nucleus may fission upon neutron absorption (symbols  $a_{ii}$ ,  $a_{ai}$ , and  $a_{i}$ ). Examples of these cross sections are found on the squares of beryllium-7, boron-10, and thorium-227, respectively.

A given nuclide might undergo two or more interactions, and its square would then contain

two or more of these cross section values. When neutron capture can lead to a metastable state as well as to the ground state, more than one value will appear beside the capture cross section for that nuclide. The cross-section value for metastable state formation is listed on the left and that for direct ground state formation on the right. For two metastable states, the higher of the two states is on the left. For example, indium-113 has an indicated capture cross section  $a_2$  of (2.8 - 5.0 - 3), which means that the cross section for formation of 44-millisecond indium-114 is 2.8 barns, the cross section for the direct formation of 50.0-day indium-114 is 5.0 barns, and the cross section for formation of 71.9-second indium-114 is 3 barns.

The designation mb or pb following the crosssection value indicates that the units of the cross section are millibarns per atom (10<sup>-27</sup> cm<sup>2</sup>/atom) or mircobarns per atom (10<sup>-30</sup> cm<sup>2</sup>/atom), respectively. When no mb or pb appears on the chart square, the units of the cross section are barns per atom.

### SPINS AND PARITIES

In the upper right corner of the square for the ground state of a nuclide, and in the upper left corner of the isomeric state, are shown the spin and parity of the corresponding energy level. Each neutron and proton has an intrinsic angular momentum of  $1_2$  (in units of h  $2\pi$ , where h is Planck's constant), similar to that of the electron, which combines with their orbital angular momentum to produce a resultant angular momentum called the nuclear spin. Since the orbital angular momentum is always zero or an integral multiple of  $h/2\pi$ , the nuclear spin (in units or h  $2\pi$ ) is always integer or half-odd-integer, depending upon whether the nucleus has an even or an odd number of nucleons. The concept of parity was introduced by the mathematical formalism of quantum theory and has no classical analogue. A system in a given state may have even parity (symbol - ) or odd parity (symbol ). For aluminum-27 the spin and parity are shown as  $5 \rightarrow$ , where the 2 in the denominator of 5.2 has been removed to improve the readability of the chart. The ground states of all eveneven nuclides are known to have spin and parity 0  $\cdot$  ; so 0  $+\,$  has been omitted.

The arguments for the assignment of spin and parity to nuclear states can be divided into two classes: strong arguments such as measuring values directly, and weak arguments such as inferring values indirectly. On the chart, the absence of parentheses indicates spins and/or parities based on strong arguments; the presence of parentheses indicates spins and or parities based on weak arguments. When the spins of both the ground state and an isomeric state are given for a particular nuclide, it is interesting to observe that these spins usually differ by two or more units of h '2-. The large angular momentum (spin) change is required for the gamma-ray transition between the states. Combining this spin change with the small energy differences (a few hundred keV) leads to a relatively long lifetime (metastable state).

### RADIOACTIVE DECAY CHAINS

As nuclear processes occur, whether in natural radioactivity or under artificially induced conditions, the nuclides change in accordance with the scheme shown in Fig. 2. To understand the use of this scheme more fully, consider the uranium-238 decay chain (one of three such chains found in nature). On the chart we start with the parent uranium-238 which emits an alpha particle. The daughter nucleus is in the second space diagonally down to the left (see Fig. 2). This square represents the isotope thorium-234. (This nuclide is also identified by the old symbol uranium  $X_1$ , which is the historic name given it before it was identified as thorium.)

Thorium-234 in turn emits a negative electron; so the loss of mass is not appreciable. However, there is a loss of one negative charge, which means that the atomic number Z increases by one. In effect, one neutron has changed into a proton. The move one space up and one space to the left (see Fig. 2) leads to protactinium-234 which has isomeric states. Each of these states undergoes negative beta emission; so another move diagonally upward to the left leads to uranium-234.

Uranium-234 emits an alpha particle ending at thorium-230. Another alpha decay yields radium-226. Three further alpha decays result first in radon-222, then in polonium-218, and finally in lead-214. However, this isotope of lead is unstable and emits a negative electron producing bismuth-214. A beta decay to polonium-214 is followed by an alpha decay to lead-210. An alternate route from bismuth-214 to lead-210 is taken in a small fraction of the disintegrations since bismuth-214 can also emit an alpha particle and the resulting thallium-210 beta-decays to lead-210.

In either case, lead-210 beta-decays to bismuth-210. Another beta decay produces polonium-210 which alpha-decays to the stable isotope lead-206. At this point the chain ends. Incidentally, in many of the above steps, gamma rays and conversion electrons are also emitted.



Fig. 2. Relative Locations of the Products of Various Nuclear Processes

Similarly, the two other natural radioactive sequences may be traced. One is the actinium series which starts with uranium-235 and ends with lead-207. The other is the thorium series, which goes from thorium-232 to lead-208. A fourth, or nep-tunium, series is also known. However, the half-life of the parent, neptunium-237, is only about two million years. Since the age of the earth is five or ten billion years, most of the neptunium-237 present when the earth was younger has already decayed, and the series is not found in nature.

Since the naturally radioactive decay chains end at stable isotopes of lead, the isotopic composition of lead ore will be variable depending upon its source and its past history. Elements such as lithium and boron also have variable compositions that are affected by reactions that their samples have previously undergone. In a similar manner, scientists examining the isotopic compositions of samples recently brought back from the moon have already obtained an estimate of the age of the samples from the relative amounts of potassium and argon-40 present. A comparison of the isotopic composition of elements on the moon with those on earth might provide scientists with some solutions to the problem of the origin of the universe.

#### Errata

#### (Chart of the Nuclides, Tenth Edition)

The absorption cross sections for oxygen and sodium should read as follows:

Oxygen  $\sigma_a$ .27 mb

Sodium  $\sigma_a$  .534

The atomic weight for praseodymium should read 140.908.

# ACKNOWLEDGMENTS

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LIST OF ELEMENTS

ATOMIC NUMBER	SYMROL	NAME	ATOMIC WEIGHT	ATOMIC NUMBER	SYMBOL	NAME	ATOMIC WEIGHT
0	n	neutron		52	Те	tellurium	127.60
1	н	hydrogen	1.00797	53	I	iodine	126.9044
2	He	helium	4.0026	54	$\mathbf{X}\mathbf{e}$	xenon	131.30
3	Li	lithium	6.940	55	Cs	cesium	132.905
4	Be	beryllium	9.0122	56	Ba	barium	137.34
5	в	boron	10.811	57	La	lanthanum	138.91
6	С	carbon	12.01115	58	Ce	cerium	140.12
7	N	nitrogen	14.0067	59	Pr	praseodymium	140.908
8	0	oxygen	15.9994	60	$\mathbf{N}d$	neodymium	144.24
9	F	fluorine	18.9984	61	Pm	promethium	· · · · · · ·
10	Ne	neon	20.179	62	Sm	samarium	150.35
11	Na	sodium	22.9898	63	Eu	europium	151.96
12	Mg	magnesium	24.305	64	$\operatorname{Gd}$	gadolinium	157.25
13	Al	aluminum	26.9815	65	ТЬ	terbium	158.924
14	Si	silicon	28.086	66	Dy	dysprosium	162.50
15	Р	phosphorus	30.9738	67	Ho	holmium	164.930
16	S	sulfur	32.064	68	Er	erbium	167.26
17	Cl	chlorine	35.453	69	Tm	thulium	168.934
18	Ar	argon	39.948	70	Yb	ytterbium	173.04
19	к	potassium	39.102	71	Lu	lutetium	174.97
20	Ca	calcium	40.08	72	$\mathbf{H}\mathbf{f}$	hafnium	178.49
21	Sc	scandium	44.956	73	Та	tantalum	180.948
22	Ti	titanium	47.90	74	W	tungsten	183.85
23	v	vanadium	50.942	75	Re	rhenium	186.2
24	Cr	chromium	51,996	76	Os	osmium	190.2
25	Mn	manganese	54.9380	77	Ir	iridium	192.2
26	Fe	iron	55.847	78	Pt	platinum	195.09
27	Co	cobalt	58.9332	79	Au	gold	196.967
28	Ni	nickel	58.71	80	Hø	mercurv	200.59
29	Cu	copper	63.546	81	T1	thallium	204.37
30	Zn	zinc	65.37	82	Pb	lead	207.19
31	Ga	gallium	69.72	83	Bi	bismuth	208.980
32	Ge	germanium	72.59	84	Po	polonium	
33	As	arsenic	74.9216	85	At	astatine	
34	Se	selenium	78.96	86	Rn	radon	
35	Br	bromine	79.904	87	Fr	francium	
36	Kr	krypton	83.80	88	Ra	radium	
37	Rb	rubidium	85.47	89	Ac	actinium	
38	Sr	strontium	87.62	90	Th	thorium	232.038
39	v	vttrium	88 905	91	Pa	protactinium	202.000
40	Z.r	zirconium	91.22	92	IJ	uranium	238.03
41	Nh	niobium	92 906	03	Np	neptunium	200.00
42	Mo	molybdenum	95.94	93	Pu	nlutonium	•••••
42	Te	technetium	55.51	95	Am	americium	
43	<b>R</b> 11	ruthenium	101.07	95	Cm	curium	
45	Rh	rhodium	102.905	90	Bk	berkelium	· · · • • •
45	Pd	nalladium	102.909	08	Cf	californium	
40	Δα	silver	107 868	90	C1 Fs	einsteinium	• • • • • •
48	Cd	cadmium	112 40	100	Fm	fermium	• • • • • •
40	In	indium	114.90	101	Ma	mendelevium	
50	Sn	tin	118.60	102	No	nchelium	• • • • • •
51	Sh	antimony	121.75	103	Le	lawrencium	
~ *		y	121.70	100			•••••
			1	1			

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#### Relative Locations of the Products of Various Nuclear Processes

		_				He <sup>3</sup>	in	a	in
		β <sup>−</sup>	out	p	in	đ	in	+	ìn
		n	out	Ori Nuc	ginal teus	n	in		
1	out	d	out	P	out	β <sup>+</sup>	out r		
a	out	He <sup>3</sup>	out		n = ne p = pr	nutron Toton		-	
					d ≠ de	auteron			

- t = triton (H<sup>3</sup>)
- a = alpha particle
- $\beta^-$  negative electron
- $\beta^+$  = positron
- < = electron capture

#### Displacements Caused by Nuclear Bombardment Reactions

α, 3n	a, 2n He <sup>3</sup> ,n	a, n		
p,n	Ρ,γ d,n <sup>3</sup> He,np	a,np t,n 3 He,p		
γ,n n,2n	Original Nucleus	d,p π,γ t,np	t,	p
γ, np	γ, P	n,p		
n , a	n,He <sup>3</sup>	Chemica Element	1) 1	
		H 1.007 <sub>6</sub> 0.33	97 — 2	- Symbol - Atomic Weight (Carbon - 12 Scale) - Thermal Neutron Absorption Cross Section in Barns
Perci Moss (Cor	ent Abundance - rbon - 12 Scale) -	Stable PGICK SATI SATI SATIS		Even Z, Even N Nuclides Have Spin and Parity O+ Symbol, Mass Number Thermal Neutron Activation Cross Section in Barns Leading to (Isomeric + Ground State) Fission Pruduct, Slow Neutron Fission of U235
Modes Rodiati in Mev, Rodiat Lived	of Decay, ion and Energy ( ) Indicate ions from Short- Doughter	Artificial Radioacti Mg 28 21.3h \$-45 (2.8) 7.032,135, .95,((78) E1.84	ly ve 3 5) .40,	Symbol, Mass Number Half – Life Disintegration Energy in Mev
	Not	urally Occurring Available but Ro	) or Other adloactive	wise
Symbol Perci Thermal N Cross Sect	i, Mass Number - ent Abundance - Hait-Life : leutron Capture . Ion in Barms			Spin and Parity Mades of Decay and Energy Mass
	Symbol –	Member of No. Radioactive De- PO218 BCL 3.0 a 6.000,5.1 B=0.020 218.000	aturally coy Chain 35m	Symbol, Moss Number Half-Life Modes of Decay and Energy in Mev in Order of Intensity Moss
Ma Ro En	Half→Life — ides of Decay, diations and ergies in Mev ladioactive Uppe	Two Isomeric S One Stabi USAN Idd IT.159 F.161 Isomer Stab	States le 61 61 73 10 10 10 10 10 10 10 10	Spin and Parity of Ground State, 1/2+ Symbol, Mass Number Parcent Abundanca Mass Fission Product, Slow Neutron Fission of U235 Isomer
R	adioactive Uppe	Two Isomerk S Both Radiooc $P_{5.3h}$ 6 $\beta$ $\gamma_{5.3h}$ 6 $\gamma_{7.3,126}$ 1212466 tr Isomer	Shates stive 3 6 5 5 5 5 5 5 5 5 5 5 5 5 5	Symbol, Mose Number Holf Lives, ? Indicates Uncertainty Modes of Decay and Energy in Mev in Order of Intensity: "Indicates Additional Low Intensity Transitions, - Indicates Several Energies Included
R	adioactive Uppe	r Isomer Rodk	Socilive Lor	~Indicates Several Energies Included

# SYMBOLS

# RADIATIONS AND DECAY

a	alpha	ogricie	

- $\beta^-$  negative electron
- β<sup>+</sup> positron
- γ gamma ray
- n neutron
- p proton
- ← electron capture
- IT isomeric transition
- D radiation delayed
- SF spontaneous fission
- E disintegration energy
- e<sup>-</sup> conversion electron

#### TIME

- ms milliseconds  $(10^{-3} s)$
- μs microseconds (10<sup>-6</sup>s)
- s seconds
- m minutes
- h hours
- d days
- y years

,

- 9 -



- 10 -

		CI		CI 32 1+	CI 333/+	3+CI 34 0+	CI 35 <sup>34</sup>	CI 36 <sup>2+</sup>	CI 37 <sup>34</sup> 24.23	CI 38 <sup>2~</sup> 0.8s 37.2m	CI 39 3/+ 55.5m	C1 40(2 <sup>-</sup> ) 0.05 1.42 m		
	17	35.453		87 9.47,47,1 172 23,477,246; - (9)	₿*45 72.93	11.46 (8*4.5 8*25, 13, 12,1330	σγ 43 σ <sub>p</sub> 45,σ <sub>q</sub> 0.1 mb	β.709, ε.β.12 <sup>πυ</sup> γ σ <sub>γ</sub> < (1)	σγ(,005+.43)	IT.66 β <sup>-</sup> 491, 11,2.8 γ2,167	β=191,218,344 γ127,25,152	β~~3.2,~ 75, γ1.46,33		
		5 29	S 30	<u>F 12.8</u> S 31.1/+	5 32	118,64-41 E5 48	<u>34,96885</u> S 34	5 35 3/t	36.96590	E4 92	E3.44 S 38	5.9 E∼8		
32.064		019s 8+	135 B*442,508	264s 8*44	95.0	0.76 0.76	4.22	87.2d β= 167	0.014	5.06m β 164,475,104	2.87h ∌=i.((4.9i,)		24	
σ <sub>1.52</sub>		γ <b>Kpł</b> 559,386,360, ·	y 677	y127	σ <sub>γ.53</sub>	σy.02 σa.16	σγ.22	10 y	σy.14	y 3 (1, 3.7)	188(2.167, 1642, )			
Р		P 28 3+	P 29 1/1	P 30 '*	P 31 "+	P 32 +	P 33	P 34 +	32.96/1	140		<u>↓</u>		
30.9738		β*11.5, γ1780,4,499,	θ* 3.95, y 128, 2.43	B+324, 223	100 σy.19	β= 1709 Poy	23.20 B-248	β=51,3.2, γ21,40						
σο 19		2.84-7.50 E14.3	E49	E42	30.97376	E 1 71	E 25	٤5						
SI 25	Si 26	SI 27%	Si 28 92.21	Si 29"* 4.70	Si 30 3.09	Si 312+	Si 32 ~650y				<b>0</b> 0			
04 25 195,347 .82 5 90	y 825	y 2 21	σy.16	σγ.3	σγ.10	γ1.27 σ <sub>y</sub> ~Ll	β=.21(1,7)) noγ				22			
· A124 **	AI 255/+	01A1 26 51	27.97693	28,97650 AI 28	29.97376 Al 29.5/t		E.21			J				
0 1295 2095 17 44 3444 1-1 3.119 8768	723s 8*324	645 74x10° y 8* 8*116	100	2.27m 8-2.85	6.52m β=25,14	73s   3.3s	18		20					
50' 7-377' 1-37,275 (1)~2 E 4 4 E   4 0	E4 26	181,112, Ε4 00	0y 234mb	¥1779	y128,243	Y 2.24, Y 2.24, 3.521 3.52	10		20					
Mg 233/1	Mg 24	Mg 2554	Mg 26	Mg 271/+	Mg 28	<u></u>	4							
A* 51, , 44	03 ay .05	oy.2	(Ty,03	9.5m 8*176,159 7.844,1015,175	21.5n β∴.45,(2.85) γ.032,135,.40,									
E 4 06	23.98504	24.98384	25.98259	αy < .04 E 2.61	.95,(178) E1.84									
Na 22 ** 2602y	Na 23 <sup>34</sup>	<sup>1177</sup> Na24 4+ 199ms 15.00h	Na 2554 595	Na 26 104s	Na27									
17 2745	0y1,40+,13}	y 2 75,	β 3.8,2.8, γ.98, 58, 40, 1.61,	А1809' В. Р.У. :										
E284	22.98977	ES 51	E38	E 9.										
0.266	9.22	37.6s	3.38m		16									
	oy.04	7.44, 1.64,	y.472D,.88		10									
F 20 <sup>2+</sup>	F 21	F 22	E2.5	L]										
11.2s #=54	4.36s 8=5.4,4.0,…	4.0s # 9.1,11.2	14											
£7.03	¥.35,1.36	E12.5	1.4											
0 1954	0 20	0 21												
8 325,46 y.20,136	β=2.7 γ1.06													
£4.82	E 3.8													
N 18 0.635	N 19													
2 47											·			
C17	J													
	12													
	t dia													





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Ar

CI

18

Ge 67 19.0m #131.2.3e y17.3413.4	Ge 68 287d	Ge 695/ 51µ5 39.2h 7085 €.8+121, 71107.574, 872, 235-204	Ge 70 20.5 97(3+30)	200msi 11d 17.023 e y.175 noy	<b>Ge 72</b> 27.4 σγ 11	<sup>5,μ</sup> ) <b>Ge73 9/1</b> 40 μs 7.8 110135 <sup>1/1</sup> 53s 72.92346	Ge 74 36.5 <sub>(15+31)</sub>	(1/1) Ge75(1/) 46s   82.8m Ir   $\beta^{-1}20.92$ , $ ^{139}_{e}$   $\frac{265,066}{618}$	<b>Ge</b> 76 7.8 σ <sub>y</sub> (10+05)	$\begin{array}{c} \text{W} : \mathbf{Ge77}(\text{W}) \\ 55 \text{s} \\ \text{g}^- 2.9, \text{g}^+ 2.2; \text{f}_6, \text{g}^- 2.3; \text{g}^-$	Ge 78 1.45h 870 7.278, 294	Ge79 <sup>β- <sup>&lt; Im</sup></sup>
60 66(0) 95h 8*4(53) 6 9(39)2752 834 4806 55(75)	Ga 67 3/ 78 2 h 7 (1933) 185 3x (1913) 888	68 2m 68 2m 107' 578	69.92425 GO 69 <sup>3/-</sup> 60.16 g.19	Ga 70 <sup>++</sup> 21 lm β <sup>+</sup> 166, γ1040,173	Ga 71 3- 39.84 a,(2+48)	Ga 72 3 37ms; 14.1h 11.100; 81.97, 4.101; 86 31,6 4.835,2.20, 630, 2901334	$\begin{array}{c} 73.9210 \\ Ga 73671 \\ 4.8h \\ 8 + 19.7 \\ 7.296.325,742. \\ (0533.0(35)) \end{array}$	Ga 74 79m 8-26,43 556,235,381 36	Ga 75 19m 9 58, 36	Ga 76 <sup>(3-1)</sup> 32s # ~6 ; 56,112,	Ga77 17s	Ga78 ~4s
Zn 65 5/ 243.7d ¢. 8* 325 yr 115, Er 349	Zn 66 27.81 <sup>a</sup> y ' 65.92605	- Zn 67 s- 9 3μ5 4 ΙΙ γ 0933 σγ 6 7 66 92 74	68.92357 Zn 68 18.57 су 10075+0 во 67.92486	137h 58m 137h 58m 11439 A 93 Par E 93	Zn 70 0.62 vy(8.2mb+89mb 69.92534	<b>397h</b> 2.4m <b>β</b> 145.8 2.61 <b>β</b> 387 21. <b>β</b> 387 21. <b></b>	Zn 72 46.5h # 30 7,145,191,016- 112 11001 # 46				46	
Cu 64 I+ I2.75h 6 573 548 8 573 570 5 8 573 5 573 5 573 5 1 1677	Cu 65 <sup>3/-</sup> 30.83 σ <sub>γ</sub> 19 64.92779	Cu66 ** 5 10m # 263,159,* 7 1039,834 7,160 £263	Cu67(V) 616h 8 40, 48, 58 7 185, 090,09 % 5 58	Cu 68(1) 31 s 8 = 3 50,2 26, 7 108, 81,1 88, 1,24, £ 4 6	CU6931 3.0m 8 - 2.66, 1007, 898, 17 - 2.03 E 2.66				44			
Ni 63 17 92 y 18 16 19 10 7 E U659	Ni 64 1.16 σ <sub>7</sub> 17 63.92796	Ni 6557 255h 9 214 610 9 1480 115, 366 9 20 6 214	Ni 66 546h 19 20 19 7 F 20 F 20	Ni 67 50s 8 41,2,32 7 90, 89,126 £41								
Co 62	Co 63 52s 9 36 9 0882	1 <sup>28</sup> Co 64 ···· 28s ··· 28s 7 095 <sup>β</sup> ·····35					42					

36 38 40

Co 54 °*	Co 55 1/2	Co 56 **	Co 57 "	+Co 58 2+	Co 59 7	21Co 60 51	Co 61 17	Co 62	Co 63	<sup>≁</sup> 'Co 6	<b>4</b> (1)			
H*45518173	1051 β*151,104,∵,€ ∵93:14/08.477	ε (1.50) ε,β*:46, 1. απ/1238 732	€ 122.014.136	11 025 # .81 48.	100	11 0586 8 318, A 155-71333,	β‴122 γ070	β <sup>-</sup> β <sup>-</sup> 2.88, 88	β <sup>−</sup> 36 x 0882	205 B	~35			
304 F8252	09/0 317 0546	-355 E457	EBS	10µ5 m, 17×10	58,93319	71333, 1173, 5720	E 129	174,2.03	F 3 6		1			
-Fe 53(**)	Fe 54	Fe 55 *-	Fe 56	Fe 57 #**	Fe 58	Fe 59 3/ -	Fe 60	Fe 61	· · · · · · · · · · · · · · · · · · ·					
2.53mi853m 5.702, j#~2.8,	5.82	2.7y ¢	91,66	2.19	0.33	45d 81 467,273/573	θ 135.137 1	6.06m #12.6,25,28,1					40	
1011 24-6,4	σγ 2 3	Гоу	σγ26	σy 2.5	$\sigma_{y+2}$	+1099,1292,192	y.0586.), 027.2 (+332,1+73,)	7120,102, 30, 12-27	i.					
2'Mp 524'	53.93962	E 232 Mn 54 3t	55.93494 Mn 55 5	56.93540 Mn 56 3t	57.9333 Mn 57	EF573	E.19	E 3.8						
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1:38 y 434,		с у.834.8	σγ 13 3	8 12 85 104,73, 9 846 (180,2113	8 26,11 7122,014 136, 22 71	y 36-2 8						30		
£47	E 598	E13/9	54.93805	£ 3702	E 2 7	E 7								
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1.5.65	7.08	a 18	Z.303	B 25	B 15				36					
	51 9405	52 94065	51 03888	1 3 64	616									
V 50∺	V 51 "-	V 52 *	V 53	V 54										
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4.53d	0.16	8.70m												
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.808 E1.98	47,9625	E 5.26	E 5.4											
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1 1.35,3.70,	Y 4.U, K B													
1 inte	L 0.00	L	1											



![](_page_188_Figure_2.jpeg)

- 14 -

42	44	46

Se 76 9.02 17.7s 17.62 17.62 17.62	77 <sup>1/-</sup> Se 78 2352 σ <sub>γ</sub> 42 σ <sub>γ</sub> (3) + 07) 8.91991 77.91731	<sup>1/-</sup> Se 797/ <sup>4</sup> 39m/≾6.5x6 <sup>3</sup> / 11.096 8 <sup>-15</sup> σ <sub>2</sub> (. <sup>6<sup>-</sup></sup> 2 <sup>0</sup> / <sub>2</sub> £.154 7	Se 80 <sup>7/μ</sup> Se 8 <sup>1/μ</sup> 49.82 57m 18m (.09+53) 13.05 β <sup>2/158</sup> 23.57 γ <sup>1/51</sup> 23.57 γ <sup>1/51</sup> 23.57 γ <sup>1/51</sup> 23.57 γ <sup>1/51</sup> 1.58	Se 82 9.19 \$\phi_1(.04+.006) \$1.91671	(V)+Se8390+ 69s 23m β 15, β 93,5, 15,2,4 52 γ10,12,0, ,356, -65,35 512,226, ε3,5	Se 84 3.2m <sup>B<sup>-</sup>1.4</sup> <sup>y.408</sup>	Se 85 <sup>395</sup>	Se86 ≥ <sup>16s</sup>	Se 87 5.8s	
54 As 75 μ- 16.4 ms 100 19μs 1024 γ 28 19μs 19μs 19μs 19μs 19μs 19μs 19μs 14.4 ms 19μs 14.4 ms 14.4 ms 19μs 14.4 ms 19μs 14.4 ms 19μs 14.4 ms 14.4 m	76 <sup>2</sup> ( <sup>N(n)</sup> As773 264h ( <b>Ι6μ</b> s) 38.8t ( <sup>172</sup> β 6 ( <sup>172</sup> β 6 ( <sup>172</sup> β 6 ( <sup>172</sup> β 6 ( <sup>173</sup> β 6) ( <sup>173</sup>	As 780 (A 1.52h P 4 - 1 y 614, 645, 151 	As79:37 As 80 11 9.0m (5s 214 - 21 - 15 6 6 6 5 3 30 - 4 5 940 2 46 - 19 7 6 6 6 78 - 2 35 2 8 6 0	As 8   3/-1 32s B <sup>+</sup> 3.8 My E 3.8	As82 15s γ.655,.817	As83 β <sup>-</sup> (n)	Δs84 5.8s (n)	As 85 2.1s (n)	As86	As87 <1.5s
Ge 74 36.5 <sup>σ</sup> <sub>7</sub> (15+31) <sup>73,92110</sup> →	75 · Ge 76 2: 9/ 2: 9/ 2: 9/ 2: 9/ 2: 9/ 78 75 75 92141	W (Ge77 - G 555 1130h 8,20 8,37,5 7,55 746,75 1,55 746,75 1,55 746,75 1,55 746,75 1,55 746,75 1,55 746,75 1,55 75 76 1,55 75 76 1,55 75 76 1,55 75 76 1,55 75 76 1,55	Ge 78 145.0 75 75, 230 He He He He He He He He He He	48		50		52		54

9 - 275,290 1829, 466 18 - 650 EL58	81.91671	1.5,2.4 3.2 y10:20.1y356, -65,35 52,226, 	y 408	р 		B			90					
57m 18m	Se 82 9.19 c.(04+.006)	(1/) Se839/) 69s : 23m 8 3.5. (8 93).5.	Se 84 3.2m	Se 85 39s	Se86 ≥ <sup>16s</sup>	Se 87			56	¥	I			
17.046 8" 44 8" / 36.977" 564 66 503.092 79 11 A09	#1.93 - y 52 (y.032,.004) (r.) E.97	8 1988 41 - 88 7 8819 14644 27 39 189 E47	β*~2 R 2 81,32 Γ2 8	8 - 3, 5, 74, y 16, 1 4, 2 P, £76	B <sup>2</sup> 26,⊷ γ14,26,10-50 (6).3	103 8~ (n) y. <sup>7</sup> 9,…	μ- in) 5	β <sup>-</sup> (n)			58			
<sup>2</sup> Br 82 <sup>5</sup>	Br 83(V) 2 40h	Br 84 6.0ml 31.8m	Br 85 <sup>67</sup>	Br 86	Br 87	Br 88	Br 89	Br 90					1	
111032 <b>45</b> 173 7-0090 7	ay(.09+.042)	87,83 8 67,11 7150 7514 11 305	æy.06	813,8,13,33 7403,2.56,85	8" 52,2.7, 7 2,4, 19),85, .028 - 2,2	<b>B</b> <sup></sup> <b>4</b> 9,4.6, <b>3.8</b> ,… <b>y</b> 22,.59,086-47	# 2.8, ·· 7.12/5, 540, 1.12, 1.538, .11 * 3.6	B <sup>~</sup> 3.6, ·· γ -51, 11	β~ (n)	β~ (n)	β <sup>−</sup>	β <sup>−</sup>	60	
Kr 83*	Kr 84 56.90	"Kr 85" 44h 1074y	Kr 86	Kr 87 76m	Kr 88 2.79h	Kr 89 3.18m	Kr 90 32.3s	Kr 91 8.6s	Kr 92	Kr 93 1.29s	Kr 94	Kr 95 Short		
249 A 89 5 249 A 89 7 19 E 89 E 248	84.91180	y108 E1.78	\$7.27 NOY \$12 E.274	y184,899,2.68, 13814,8 σμ10 Ε5.2	γ203,125,165 2.20,155 3.5 E4.49	58,22, 1783,53 52 56,5	y 094, 35	(n) E8	(in)	(n)	(n)	(n)	(n)	
**Rb 84 ? 20m 33.0d	Rb 85 <sup>%-</sup> 72.15 ¢(.06+40)	1 Rb 862 1.00m(18.66d 11 56 18178, 74	27,85 5.0x 10 <sup>10</sup> v	Rb 88 <sup>2~</sup> 17.7m #`52,33,2	Rb 89 15.2m 8 <sup>-1</sup> 26,2.21,4.49	Rb 90 4.28ml 2.7m β⁻ β⁻66,	Rb 91 57.9s	Rb 92	Rb 93	Rb 94	Rb 95	Rb96 0.23s	Rb97 .14s	
17 25" . K.+15 (E):	65,90929		87.90564	0,05 E1463	σ <sub>γ</sub> 0.9 E 546	y (551),65-1 65 E2 67	E19	E4.8	E3.5					
70m 64.7d 11008 6 23 y 540	9.87 57 (.84 + ?)	2.83h 7.04	82.53 «,8mb	508d 8 146, ~	28.9 y Ø=546.(2.27)	9.67h #109,136, 267	2.69h 8	7.5m #~2.9,26,2.2	1.29m #*2.1	26s #-	β <sup>−</sup> 4.0s	~ 0.4s		
Sr85 *	Sr 86	Sr 87**	Sr 88	Sr 895	Sr 90	Sr 91 54	E289 Sr 92	50 Sr 93	Sr 94	Sr 95	sr96	Sr97		ļ
170-0-€,8*:2, - β-20-8, 6:3 - β	11 38 €,∄*8 1¢ γ 48,(39) 1∂*154	11 39 €.8*78 15.4ms (+184, 11 25 90,	e <sup>-</sup> 1.28)	IT 48 8 2.2" y.20 y.75 B .62 0y < 6.4	(T.55) β <sup>-1</sup> 54. γ   2) α,ι 4	8-363,132, 159-271 793,47,1.4,	B-2.89, y.27, 94, 49-2.	β <sup>∼</sup> 5.0, · · 4 y.92, 55, 1.14,1.67 3.5	8~4.3,76,117,~ 7,95,2.18,132, .43-4.07	β~3.5 γ.7,10,··	₿ <sup>-</sup>			ļ
8+ Y 864 48m 14.6h	13h 80h	0.30ms 06.6d	15.7s 100	(7+)Y 90 2- 3.19h 64.07	9/4 Y 91 1/ 50.5m 58.8d	Y 92 2 3.53 h	Y 93	Y 94 20.3m	Y 95	Y 96 23m	Y97		Y99 ∼ 0.8s	Γ
7 38) 63 50	> 390 e <sup>-</sup>	24 1 15 E2 83	7218 8990470	90.90564	91,90503	{y:030} σ <sub>y</sub> < 4 E.09	93.90631	(.160) y.756,.723,(.23, E1,121 .765]	95,90829	y.11-1.85(.743) E2.67	E~1.5	E4.5		E~6
8* 210.	f 850	4.20m (84h	0.80s   51.46 172 319, 07 07	il.23 ¢,0.9	17,11 a,0.18	9.5 x 10 <sup>5</sup> y #~.063, 034	17,40 c.05	65.5d 8*.40, 36,89,	2.80 5,0.2	16.8h ∦≊19i,…,(i,27;	31s	2.4s 8-	$\sim$ is	β-~~

![](_page_190_Figure_1.jpeg)

![](_page_190_Figure_2.jpeg)

$\begin{array}{c c} 4.7h & 15.0d & 21h \\ \epsilon_{,B}^{+,\gamma_{1,\alpha}}, B^{+,\gamma_{2,\alpha}}, 5\epsilon_{,B}^{+,\gamma_{2,\alpha}}, 6\epsilon_{,B}^{+,\gamma_{2,\alpha}}, 6\epsilon_{,B}$	4476 <b>3y</b> 2 ty 207 €,17158€ 7 1 1 58 € 7 1 1 1 58 €	3 55m 100 436m 429 1T ∪40 7(11 + 137) 102×054 (02×0554 102 104 104 104 104 104 104 104 104 104 104	s 38s 35.5h 2.48h 30.0 7 24 7 24 7 37 38 35.5h 26.47 56.75 47 56.75 57 7 38 38 565 56 57 57 70 75 57 7 56 56 58 56 7 56 56 58 56 56 56 56 56 56 56 56 56 56 56 56 56	\$ 22.4m  7s <sup>4</sup> β <sup>−</sup> (2,84−15) γ 31,12 <sup>−</sup> 68 y 43,62	$\begin{array}{c c c c c c c c c c c c c c c c c c c $	
Ru 98 Ru 99 <sup>54</sup> 1.87 12.72 4,<8 4,4	Ru 100 12.62 4,58 7,5 7,5 8	™         Ru 102         Ru 103' 31.61         39.8d β <sup>+2</sup> 39.8d 31.61           g <sup>+3</sup> g <sup>4</sup> <th>E bec         E3.67         E3.87         E3.89           5<sup>th</sup>         Ru IO4         Ru IO5<sup>th</sup>         Ru IO5<sup>th</sup>           18.58         4.44h         8<sup>th</sup> (5.161,03)         975 (26th (7.161,03))           97,5         97 (25th (7.161,03))         97 (25th (7.161,03))         97 (25th (7.161,03))</th> <th>β         368d         4.2m           β         394.354.1         β         3.001           γ         9.1.67.3001         γ         9.1.13</th> <th>Ru 108 Ru 109 45m 35s 8<sup>-13,12</sup></th> <th>66</th>	E bec         E3.67         E3.87         E3.89           5 <sup>th</sup> Ru IO4         Ru IO5 <sup>th</sup> Ru IO5 <sup>th</sup> 18.58         4.44h         8 <sup>th</sup> (5.161,03)         975 (26th (7.161,03))           97,5         97 (25th (7.161,03))         97 (25th (7.161,03))         97 (25th (7.161,03))	β         368d         4.2m           β         394.354.1         β         3.001           γ         9.1.67.3001         γ         9.1.13	Ru 108 Ru 109 45m 35s 8 <sup>-13,12</sup>	66
97,90329 98,90394 TC97 30 TC 98 900 2 6 × 10 <sup>6</sup> y ~ 15 × 10 <sup>6</sup> y 1 <sup>1</sup> y 7 5 6 66 7 75 66	99,90422 100,90338 1 TC 9947 TC 100 50070,213,105 y 1595 11 8:292 8:34,25 42 70,22 9:54,6 42 70,2 10 4:50 4:50	$\begin{array}{c c c c c c c c c c c c c c c c c c c $	103,90543 ЕГ.87 Тс.103 Тс.104 в 22.70 4 715, 35, 275 18.0m 6 724, 7 36, 53, 89.16 6 4 7 7 5	E0394 TC 105 7.8m 8° 34,18 34,18 34,16 34,16 59,105 75 75 75 75 75 75 75 75 75 7	Tc 107 Tc 108 295 < 105	
$\begin{array}{c c} M_0 & 96 \\ \hline M_0 & 96 \\ \hline 16.53 \\ \sigma_{\gamma^2} \\ g_{5,90467} \\ g_{5,90602} \\ g_{5,90602} \\ \end{array}$	MO 98 23.78 σ <sub>y</sub> .14 97.90541 4.19 4.	Mo         IOO         Mo         IOI           9.63         14 6m         14 6m         14 cm           q, 20         β <sup>-</sup> 2 23, ·         y102, 59 2 08         98-166           99,90747         E2 82         98-167         14 cm	$\begin{array}{c c} & \text{MolO3} \\ \hline \text{MolO4} \\ 11, \text{im} \\ \beta^{-1} & \text{if} (-4) \\ E & 1 \\ \end{array} \begin{array}{c} & \text{Sh}^{2} & \text{Gos} \\ & \text{Sh}^{2} & \text{Sh}^{2} \\ & \text{Sh}^{2} \\ & \text{Sh}^{2} & \text{Sh}^{2} \\ & S$	Mo IO4 1.3 m # 12,4 5 # 1070 1.56,86,3 Mo IO5 4 15 #	MolO6 9.5s	
(1) Nb95 <sup>1944</sup> Nb96 87h 35.1d 17 235 β 1600 β <sup>2</sup> 746 5100 g 75 7 16 16 15 925 E3 (5	Nb97 (92) 58s 736m 2.8s 51m 2.8s 51m 2.656 736m 2.8s 51m 2.656 736 1437 2.656 746 1437 2.576 1437 2.687 1437 2.697 1437 1437 2.697 1437 1437 1437 1437 1437 1437 1437 143	Nb 99 Nb 100 145 24m 663 29m 145 75 75 75 75 75 75 75 75 75 75 75 75 75	Nb 101 7.0s 		64	
Zr 94 Zr 95 <sup>5</sup> / <sub>6</sub> 17.40 65.5d g.05 (6 <sup>2</sup> 40, 36,89, (160) 756,723,123, 93,90631 E1.12 765	Zr 96 Zr 97 2.80 16.8h 57 0.2 7 10. (127 7 10. (127 7 10. (127 7 10. (127 7 10. (127 7 10. (127	Zr98 315 B <sup>-</sup> E~1.5 E4.5		62		
54	56	58	60			

Rh 99 47h 15.0d 68*14.8*3	Rh 100 21h 6.8+2.5	™ RhIOF™ 4470 3y €17158[€	Rh 102 2 ly 2078	<sup>7*</sup> Rh 103 <sup>µ</sup> 55m 100 I⊺∪40[q(11+	Rh 1041 436m 42s 8 247	1/2 Rh1057 385 35.5h 17 (29 6 566 55	Rh 1061 2.18h 30.0s 70 8 354	RhIO7 <sup>15/1</sup> 22.4m 8 <sup>-1</sup> 2.84-151	Rh 1081 175 8: 455554	Rh 109 ~50si~30s	Rh 110 <sup>1+</sup> 5s 8 <sup>-</sup> 55.	
Ru 98 1.87 4.5	Ru 9954	Ru 100 12.62 4.5.8	Ru 10 M	Ru 102 31.61	<sup>2</sup> , ax <sup>b2</sup> E247 Ru 103 <sup>56</sup> 39 Rd β → 22	Ru 104 18.58	Ru 105 <sup>54</sup> 4 44h	E:51 Ru 106 368d #= (394, 354, -	Ru 107 4 2m β 32.00 30	Ru 108 45m β=13,12	Ru 109 35s	66
97.90529 Tc97 ** 90d 2.6x10 <sup>6</sup> y	<b>98.9059</b> 4 Tc 98 ~15 x 10 <sup>6</sup> y	99.90422 TC 994 60070 k215x103	100.90558 Tc 1001 15.95	101.90435	E	103.90643 Tc 103 50s	E187 TC 104 18.0m	(γ 5) 2, 62 3 (6) σ <sub>5</sub> 2: E <sup>0</sup> 0394 Tc 105 7.8m	E3: TC 106 375	E.3 Tc107 295	Tc 108	00
Mo 96	MO 97 M	Mo 98	Mo 99 ·	Mo IOO	Mo 101 **	E2 4 Mo 102	7 36, 53, 88, 63, 6147 159 MolO3 5,352 665	143, 159, 108, .321, E34 Mo 104	Mo 105	MolO6		
st.90467 \$5,90467 ⊡Nb95!94	σ <sub>y</sub> 2 <u>96.90602</u> Nb 96	97,90541	B=123, 44, y(140,2, 74, 31 = 23 E137 Nb 98	99,90747 Nh 99	β-223, 902,59208, 08-166 E282 ND 100	β <sup>-</sup> - 1(r - 4) E - 1 Nb 101	6 6 594, 26, 17 3 142	1,3 m β 122,4 5 γ 170 1.36,88,3	8	7.75		

62	64	66
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	H 12, 84 51	H	11' H . 41 S	1/5 N.1	t		68		70		72		
1 . 3.	1	l' · · ·		[		1					. An		
	م دام	4			4	1							

- 18 -

								54	Xe 131.30	Xe115	Xell6 55s	Xell7 65s	Xe118 6m €,8'	Xe 119 6m €.8⁺
								J4	σ <u>a 25</u>		Y 104	Y 059, 117, 222,	y 053, 117, ·	r 098.23i.
									Ι		1115 13m	I  6 < 5m	I   7 24m	I   8 9m  42m  8+6.4
								53	126.9044 <sub>06.2</sub>		β'	в' у 104	γ i6, 33, 72, i	y 60, 51 y 55,605 E ~ 7
			Te	Te 107	Te 108	Te1092 4.2s		Tell12 19.3s			Te   4	Te 115 0.10s 6m	Te 116 2.5h	Te 117 ··· 1 64m
		52	127.60	₿* 0328	β* ε α 3.08 (p)3.4, 3.7, 2.6	(g)		(p)			7(89,130)	21, 6 772,128, 138,96 645, 22	y .094, 11.29 93,.97,2.22, 1 F16	7 720,1 12, 09, 57 - 2 30 E 3 50
			Sb							Sb 112 0.9m	Sb 113 6.7m	Sb 114 3.4 m	Sb 1155/* 31.9m	© 56116 60.4m _16 m
		51	121.75							β†.∉ γ1.27*	(8*242,+85 € y 3-+5,t0791	β <sup>+</sup> 4-0,2-7 y 89,1-30	€,8 <sup>+</sup> 15: y 499,114*222 €3.03	68 6 7 3 712996
			Sn				Sn 108	Sn 109	Sn 110	Sn 111	Sn 112	""Sn113"	Sn 114	Sn 115"*
		50	118.69				* 78, 42	β+ ≼,β*~16 γ 34.12 891.66	€ (∦*2.25) y 283,(66)	<ul> <li>ε, β+1 51</li> <li>x 115,0 76,1 91</li> <li>373-2 32</li> </ul>	σy(3+8)	IT 079 € e y 255. € (392)		2 5. 1149033 - 3µ5
			σ₀63 In			In 106	In 107	In 108	***InI09**	149b 67m	<sup>111,9048</sup> In 1119/*	4'In 112 *	"In 11394	" [n   4" 44ms 719s
		49	114.82			β*31,49 γ165,185	B*23, € , 22, 28 - 23	β <sup>+</sup> 3.5, €, β <sup>+</sup> + 3 2.7, γ 24, 15, € γ 63, 84μ <sup>8</sup> + 105	11.68 (.8* 80 71.43, , 20, 63, 17:004(.33)	€ 8+2.25 y i2, 66, €, y 66, 1 i1= 94	11.54 € 17.54 € 17.3, 17.3,	IT '56   B 66 ei6, B* 157 y 62, 69 1 5	17 392 0 (2.8+ 50+3)	$\frac{\gamma - 31}{5} = \frac{\beta^{-1} - 98}{30}$ $\frac{5}{5} = \frac{\beta^{-1} - 98}{5}$ $\frac{\beta^{-1} - 98}{10}$ $\frac{\beta^{-1} - 98}{10}$
	Cd	CdIOI	Gd 102	Cd 103	Cd 104	Cd 105	Cd IQ6	38 E4 5 Cd 1075/4		<sup>10</sup> 11 (E 3 93) Cd 10954 12 (c) 4534		E 66 E*259	Cd 112 24.07	<sup>772</sup> 56 E <sup>+</sup> 43
<b>48</b>	112.40	<b>#</b> *	β+ γ 118, 481,104,	β+ γ108,145,146	€ (β*2.7) γ.084, ∪67, , (556) €	€,8*169, 708,35,168, 131,	τ.ε.ε σ <sub>γ</sub> ι	€,8 <sup>+</sup> 30 γ(093),033 122	σγιι	17 060(¢ (y 088.) (y 700	σγ(.1+11)	y 247D 05 24	σ <sub>y</sub> (03+2.2)	β <sup>-58</sup> 1T 27 σy2x10 <sup>4</sup>
	σο 2460	L	L		E~12	E~28	105.90846	E   4 7	107,90419	E 160	109.90301		111.90276	
	52		54		56		<b>58</b>		60		62		64	

Cs 132.905

σ<sub>0</sub>29.9

55

Cs120

- 19 -
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(13 903M	146 141 E163 F145	115,90478		70	END .	70		74		10			
Cd 114 28,86	"Cd115 4410,5355 8 442,2210	Cd 116 758	Cd 117 **	Cd118 49m 6 4-1 6 4-1	Cd 119		Cd  2  - 13s			76			
14 5110 4 1901-65147 114.901387	11.64/ P.33 P. 1541m E.33 A.104.6765 2.2914.189 2.117	1.77 ρ.74 1621 - 56 μ. 1.51 1.51 1.51 1.51	6 3, 8 49 20 201 12 20 347	Η 27 Γ - R	1019 P6.	, 11 a 	8°~5 1√114,10 1∼67	R 45 P	A 53 y113, 99,321 £~74	105 2.05		78	
©In 1159"* 450n 9572	1ª 'In 116 '*   12 35    142 511	In 117	In 119 ** 1 15m 5 is 1	"In 119"" 18m - 2.2m	Tin 120	Jn [2] 31m - 30s	In 122	In 123	In 124	In 125	<u> </u> E~31	EI 5	ļ
σ <sub>y</sub> (106+2)	111°9 πy3 y 16 #6.90296	σ <sub>γ</sub> (65+²)	11.57 K 5.550 my 2 \$18.90*KI	αγ(~ αα + 16) Πα αργγα	1 240 ju 7324 151 min	071001+151	12 - 24 - 18 - 42 9 166 - 9 1 08	(Ty ( 14+4 min)	8 204 8 234 7 332 7102915 28 195 23 228	B <sup>-</sup> (1,9) 7092,067,060 (42,67,-)	B"27 B y 49 y 10, 82 27 29	β=.8, .7, (2.5, ··) γ 04~ 57, (75, 31, ··]	
Sn 116	Sn 117 "	Sn 118	" Sn 11914 2451 - <b>85</b> 8	Sn 120	1-150121 1-760 10621	Sn 122	1403ml234	Sn 124	19:5n 125m 19:6m 12:654	Sn 126	Sn 127	Sn 128	
7 16 1 82	4 49,124 4 4 40,5 07 19123 EA	(1247 38	(AN) A (AN) A (A	σγ(D6+2+2)Ω 	the man provi		06,6 7,60,69 11,001,54,27 1,50,65,7,2916	428 6(X), (09) 67(109, 035) 8,76	ν 42, 67 - 42, 62 11 2 Ε3.7	(69)) 7686,473,78,25 (42)10061- E160) (29)	y 75, 31 y 31- 75	(1 45, -) 7 8L54,92,18-2.1 ( 027 - 140 ) L∼2 5	
Sb 117	Sb118	Sb 119 38 lb	* 55120 + 5.164 - 16.0m	Sb 12154 57,25	. St.122'	Sb123** 42.75	Sb 124 20.3m 60.2d	Sb 125 <sup>54</sup> 2.73y	Sb 126	Sb127	Sb 128 10.4m 9.0h	Sb 129 4.34h	
(1123,00)	15 219 27 179 1029 (1224) 72293	のyi 34+2(1) いり 2010 (	vizi) vi67 Kg Kigli jELi29	σμίτι) Οι 5.107	1	0~(05+7) (23.00244	2 0350 03 L6 2490442	07113+91 125.99332	87. y 174236. 06. E 69	σy (.016 + 20) 127,90448	e-, 8- 17028, 45, 16,91, 170, 746,02 21-126 7-140, E1 48	65y (.02+.20) 129,90524	
6004	10 119 16h	1e 120 0.089	16121 1504 - 131	Je 122 2.46	0A7	fe 124 461	Te 125** 58d 6 99	Te 126	109d 93h 109d 93h 109ez 8169	Te 128 31.79	Te 129 34 34 ld 69 m 11.095 8 14510	Te 130	
1.15H,0E3100	106, 54 12, 44	40,350 291,184 11,14	E414	F 51.4	1 - 140, 40, 70, 1 - 44 - 2 - 7 5 - 4 1 - 74 - 2 - 7	1911-55 1772-1468 11-120	2 10 41 20. Print 45. E 125 E 7350	126.914AT	4.8+ Y 743 E 214 E*(2*	17(140) e- 07y(18+9) E.19	γ 54, γ 67.54.74 σγ 19 Ε2.99	γ.364,0800- 723 Ε.970 <sup>-</sup> 723	
20) m	1 12∪ 53m 135hΩ β*39 ∉ θ*15 1	1 1212h 1212h 11 4.8+12	6 166 35m 8*30 256, 6	1360° 1316 1	1 1754 0 174 1.81155,215	1 120 " *97d	1 140 ° 1304 6767,74142 2° 38 125	1 12/3	25.01m	1.5 x 123"' 1.5 x 10 <sup>7</sup> y 1 <sup>8 - 15</sup>	1 1.70 89m 12.4h 11:048/8=62.104	8.065d 8.065d 8 <sup>-</sup> .606.25-8i	
TIIQ	44 13 8	1 101	F2.8 7 100	123 20F		:25.90429 T 1:05 5/4	5.54	127.90354	1299047	129.90351	3090505	60, 18 13190446	
40m 9* • 151, 073, 175	39m 6*78,4 7096-080, 32,	20.05 «(#* 1)2 «(#* 1)2	12 () F 1, K * 17 1, 149 170 130	0.005	1974; 1974; 1974; 1984; 1984; 1984; 1977; 1984; 1984; 1984;	(0.090) (0.090)	7018 36,418	1.92 Ty(.4+<8)	Rd 25.44	4,08 0y(4+<26)	11.96d 21.18	8.3 ms 26.89 7773. 67.54 ox 03+4	
Xe 120	Xe 121	Xe i22	Xe 123	Xe  24	Xel25	Xe 126	<u> ⊧393</u>  ™Xel27™	Xe 128	Xe 129	E36 Xe 130	E*208 E 12 ₩Xe I3I 3/+	132,9054 Xe 132	(
	,	0** 21		e a: 24.	114 x	27 4167 1.17 12 40 1.15, 469, 130 1.38	18+2912AR 8 443,526 296	Y 27 AL SE .	)#,8*1 97  x 536, 596, 894   12-2 15  8-442	e nay	6.8* 4 7 669.363-1985 8 7 7 465 567 1031	σ <sub>γ</sub> (2.7 + 27.2)	
CS121 2m	0.51227	US 123 55m	CSD/4	- GS 1, 154 - 454	- Cs1//5 **	- US 12711 - 5,25	US 128'' 38m	US 129" 32.3h	Cs 130"	069d	CS 132	CS 133"	

° (0 29)	5 134 <sup>41</sup>	y 53ml2 3x10 <sup>6</sup>	<sup>μ</sup> t Cs 136 <sup>5</sup> <sup>5</sup> y 13d <sup>β M βF</sup> <sup>β M βF</sup> <sup>β N βF</sup>	Cs 137 <sup>#</sup> 30.2y <sup>(c1 t)(4)(176)</sup> (c1 t) (c1 t) (c1 t) (c1 t) (c1 t)	Cs 138 32.2m (24.54) (143.00,46) (222.00-337	Cs 139 9.3m # 42~? 7.~3 y 128, 63,	Cs 140 6385 81544 759, PR-315	Cs  4  24 7s (n)	Cs  42 1.7s	Cs  43 1.7s	Cs  44 e <sup>- 1.1s</sup>	
1 2 24 L' 7'	3 133 V 3 133 V 3 2 344 y 0410 17 190 16 428	Xe 34 3 305 10 00 10 24 10 00 10 24 10 00 10 24 10 00 10 00 10 00 10 00	Xo 135	Xe 136	Xe137	Xe 138 14 2 m 5 P2 2 M (15m 2 42 2 42 2 42 2 42	Xe 139 40 a a - 48, v 219, 075, 295- 146	Xe  40 13 6s	Xe  4  1.72s 8" In)	Xe-142 122s <sup>β<sup>-</sup></sup>	Xe 143 096s	Xe 144 9s #-
I 2. 8. AC 7.67, 15	132 4+ 284h 2-2 44 17, 52, 2 7 E3564	I 1337 208h 81 (23)54 53) 87 ( ),	74 I 134 52.3m 8.22.3m 7.43.3.14 2. 7.447.884 14.12 F4.2	I 135 67h 67h 67h 67h 67h 67h 67h 67h 67h 67h	1 136 <sup>12</sup> 855 8143,55,27 70,7 7132,20-32 870	I 137 22.3s β <sup>-</sup> (n) 6	I 138 60s (n)	I 139 2.0s	I  40 1.5s β <sup>-</sup>			90
1 Ti 30 8 4 1 Tile	a 131 34 h 125.0m 1 25.0m 1	Tel32 786 2009-276 2009-276 2009-276 2009-276 2009-271	53m 125m 63m 125m 8 32 alerso 6 ana ang 6 2 ang ang 6 ang ang ang ang 6 ang ang ang ang 6 ang ang ang ang ang 6 ang ang ang ang ang ang 6 ang	19134 437	Te 135	Te136 A <sup>1: 33s</sup>				88	***	-
Ял , 20	3130 1 1 37m 12 , 19 12 , 19	Sb131 23.0m 7 95, 64	Sb132 45s 1,3 im	55137 2.7 m 1 - 51210253 1 - 51210253	Sb134 11.35 (n:	Sb135 <sub>д</sub> 1,708 д						
Si P 2m	n 129 1 - 7.5m 1 - 7.5m	Sn 130 26m	Sn 131 1.3m	5 <u>Sni22</u> 10n	Sn133 ~ 55s #			86	4			
h4		80	a <sub>waan</sub>	82	2	84						

![](_page_194_Figure_0.jpeg)

Pm 61

Nd137 38m

Pr 136 13.5m

€, β\*

Nd136

Nd135 12m y 21, 44

Pr 134 17 m β<sup>+</sup>.ε γ 41, 64, 22-

Nd

144.24

σο 50

60

Pr

140.909

		78		80		82		84		86		88		90
61		∼ hm β' ™	⊃ 8 m €, 8* 70 05, 43, 78)	209m #*~26,4 ;:22,89,195 207 E36	365 A*38;-:+ y157 E4.8	2650 € 1 7 742 • E(Q8	3630 € + 6(8, 697, 477, 302 - 15) E2 37	189 5 072, 067D 02 24 E 14	') 5 3 y ε <sub>1</sub> y 454, 736, β <sup>-</sup> 79,y 747, <sup>n0</sup> έ <sup>+</sup> σγ 8400 Ε <sup></sup> 154 Ε <sup>*</sup> 149	2.62.34y β= 225 γ.12.0 σγ.(91+102) Ε.225	6 597 8 247 683 495, -02, 47 90% - 550, -02, 47 90% - 550, -12, 47 11.0615 - 42, -12, 42 2, -8200 - 5, -12	33.1N β=1.07, 78, γ 286 = 85 σ <sub>γ</sub> 1350 EL071	2 0011 8' 2 3,1'8, 4,3 5, 7 33,117,1 33, 83, 175, 41= 3 1 2 3 4	20 4 Γ β=85 4 3 24 3 7340,026 9ō ε σγ<700 ε γ
	Ρm	Pm139?	Pm140	Pm 141	Pm 142*	Pml435	Pml44	Pm 145	Pm146	Pm 147"	**Pm 148	Pm 1497+	Pm 150	Pm 1515/
	og 5860			E21	15%6,1:73, 273-217 E 3 49	143,9120	σ, <80 E.63	145.9130	σy € 5 .46.9149	147.9148	σ <sub>7</sub> 41,800 σ <sub>2</sub> 05 148.9172	149.9173	σχ 5.200 Ε. 75	151.9198
62	150.35	14m •	2.5m + (A*~2.6)	725m F,8*10138	16:35 883m 117:50+.8*247 21056	3.09 <sup>o</sup> v~ 7	5400 F	1.0 × 10 y	14.97 107 x 10 <sup>11</sup> y 12.23	>3x 10 <sup>14</sup>	>13.03 >1x10 <sup>15</sup> y	σ <sub>γ 102</sub>	2000 2007	Ty 208
	Sm	Sml40	Sm 141	Sm 142	Sm 143 -	Sm 144	Sm 145	Sm 146	Sml47**	Sml48	Sml49*-	Sm 150	Sm151	Sm 152
	00	9 4 2 x 10 <sup>3</sup>	a an	11-1,-4, 150	5 82 3266244 E.6.38	7 894,166, 01 7 51 E 2 72	7 147, 633, 666 703, 145, 2.16 1, 3, 87	145 145 161.77	7 510, 61 65 098 65 01 6 64 6 4	497, 023 5# E - 4	446. +334.467 36.164 -209 -6 	1 510С (60 элчн	на и с 102 и с 102 и с 1410 Г. 508 м.с.	152 9214
	63	151.96	L m	2.6m #*41	10s 8*5	5.96d	<b>4.6 d</b> €_8*14780,2 II	24.3d	54.5d	24/µs 93d 5 860, 4 7 5 934 22	6 Cy 12 6 h €£1 1 √34, 2-8125	60µs 4782	Tria Arti e di Tria Arison Sulta Angli al	52.18
		E.	Fu  42"	Eu 143""	Eu 144	Eu 14515 *	E2 22 Eul46**	Eul47919:	Eu148 <sup>65</sup>	149 9186 11/-Eu 14915/1	Eul50	Eul51 5#	Eu 152	Eu153 5
		04	σ_46×10 <sup>4</sup>	ľ	y 176,188, 33- 1.6''	7 22 - 7 116. 155	129, 196, 128, 170, 107,133		185-108		y 6220,054,175, 1 35 a 2.6	a 2.14 0 = 190	0697. 0974	
		C A	157.25	45m #*	22,9m €.#*25	2 9h' 50d	38h € 8*97+25	93y	9,54	1.8x10 <sup>6</sup> y	1200	0.20 El x10 <sup>14</sup> y	76µs 242d	2.15 5,60
			Cd	<u>कर</u> Gतावय	Gd145	Gd 46	Gd 1472	Gd148	Gd 149 <sup>77</sup>	Gd150	Gd15117	Gd 152	Gd 1531	Gd 154
			CO			n ng Skoo	78,112	a399 y 7-12	y 64, 93 0	γ 108,15 ·13 α 1 4 2	23 344,587 a	9088.212 07016 36	y (23,	+ 0866, 1, *3, - 019 - 61- - e1
			65	1 b 158924		100m 24m	70m €.8*4.6,	415m, 4.13h	3.in €. <sup>β+</sup>	129h	4m  76h €,8* €,8 2,8,19,	0.19ms-24d	8h 18 h	5.4d
					$\sigma_0^{0.00}$	THUT	Thias	Thida	E-2.9	1519247 Th151	152.926 Th 152	1.53924 Thi53 <sup>54</sup>	Tb 154	55.924 Tb 55'3'
				<b>66</b>	162.50		ŕ	8*. 0.42	β*.« α 406	γ.256 α37	y 381,400, 182 54 α≛5	a 5.35 a 2.85	A552084-104 Na HV F	2 ×10" •
				~ ~	Dy		Dy 149 ∼15m	Dy 150	Dy 151	Dy 152	Dy 153	0y 154 ∣3h ~10 <sup>6</sup> y	Dy 155" 10.2 h	0.052
						σα 65			E 6.4	0.000	1.25	e-	D ICC/141	DUIEC
					67	164.930	√ 20s	a 4 60 a 4 51	a 4.45 a 4.38	a 400 € a 395	a'.c [a3.9] 7,335,412, 72,15-	β*or€ <u>β</u> *21 α396 χ040,	€.8 • [8,2,9,÷ 3 7 138, 266, 366	β* 2 ∩87 1.2 1
					<b>.</b>	Но	Ho 150	Ho 151	Ho 152 525 12.5m	Ho 153	Hol54 3,25m (118m	Ho 155 2165m: 47m	Ho 156	Ho 157
							Ho150	Ho 151	Ho 152	Ho 153	Hol54	Ho 155	Ho 156	H

	69	Tm ا68.9 <b>34</b> م <sup>113</sup>	Tm153 158s ¤ 511	Tm154 2.98s 1 5s 2504 2496							Tm161 30m \$ 046373	Tm 162 79m 22m * 102, 4*23, 24 38,9 * 102,	Tm163'4 (,8h €,8+13,4 y:104,242,022- :.80 E2.27	Tm164(+) 1.9m 8+2.94(-),* 7.09(,2)=7.38 5.3.96
68	Er 167.26 		Er 152 115 a 4 HO #*	Er 153 365 9 4 67	Er 154 5m ¤ 415	Er 155 5.3m @ 4 0	Er 156 < 4 m	Er 157 24m #* y.117-200	Er 158 2.4h #+ B r(067),072:.96	Er 159 36m (206),37- 260	Er 160 29h ¢. (8+57, ) (y 729, 966, (y 729, 966, 087-2 8)	Er 161 *** 3.1h €.#**2.8k. 7210 826.06* ***	Er 162 0.136 0,136	Er 163 <sup>5/-</sup> 75m • 43.1(301,
67	Ho 164.930	Ho [50 , 20s	Ho 151 42s 36s a 460 a 451	Ho 152 525 2.5m a4 45 a4 38 E 6 4	Ho 153 6.5m 93m a 400 6 a 395	H0154 3.25m [118m #*,e a3 91 y.335,412, 477,158 1.25	Ho 155 216.5m 47m $\beta^+ \text{ or } \epsilon = \beta^+ 2 + \alpha^- 3 - 36 = 0.040, 0$	Ho 156 55m •	Ho 157 15m 7 0H7 1.20	(2-)H0 158 (5*) 24m 11 3m 4.61 3(4,8*)30,185, 7.09 <sup>3</sup> 4.093, 218, 1.07 32 2.62 1.67 32 2.62 1.67 52 2.62	Ho 159 8.2s 33m 17 206 5 7 031: 395 Ev 1.6	12-71 Ho 160 <sup>(51)</sup> 4 7h 25m 11 060 <sup>(6,01-57)</sup> 4.2 <sup>+</sup> 7 <sup>729,966</sup> 4.2 <sup>+</sup> 7 <sup>729,966</sup> 911-2 8 087-2 8	14H0 161 7/- 6.8sj 2.5h 17 211 k e y.026, 10, 078,044- 76 ; E ~.8	
	82		84		86		88		90		92		94	

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,2 1Ho 158:51	Ho 159	<sup>21</sup> Ho 160 <sup>61</sup>	4Ho 1617	*'Ho 162 ''	11-Hol63	Ho 164	Ho 1657	10Ho 1660-	Ho 167/7	Ho 168	Ho 169 <sup>77-1</sup>	Ho 170		
24m j 1) 5m 1,813 (8,23,185	8.2s! *3m	47h 25m	6.83 2.5h	67m 13m	:07s 1~33y	36m 2	100	1.2×103/26.8h	3.1 h 8:32,97,61	3.0m	4.7 m	44s		
099 099 218	y 01-245	14,5" 7.779.966.	P y 026, 10 078,044	7.058 Y 081	1400 J	Y073	oy (3.2+62)	70806, 175, 184, 811, 70806, 380	2080,.074-	7.85	y.78, 85, 06 .92	y.43		
£ 4 22	E-ULB	jr 3 3	1.76 E = B	5.081-1227	16.009	E 103 E1 11	164.9304	2ims 184-183	E 0.97		EZI	E 4.2	ļ	
÷ Dy I57∞	Dy 158	1-Dy 1593/-	Dyleo	Dy 161 5/+	1 Dy 162	Dy 163*-	Dy 164	Dy 165 /// 32st 2.35h	Dy 166	Dy 167				
\$ 326.82.082	ey100	11 15 COA 080	1. E.	7.590	#2200	σ.140	σ(H2C + 7BO)	17 2 B + 291 25,- 7 J + 094 * 362, 11 1 261 + 1297	B40,.48,	β- 19-57			104	
1 (60,144,1796) 10 - 1	1579244	22 8	159 9252	14.0360	161 326 8	162 92 9 9	163,9292	17508 7510 8 5 7510	40	/51				
PTb 156	Tb157**	1.6168,64	Tb159**	Tb160	Tb I6I 3/+	Tb162	Tb16313#	Tb164	<u> </u>		t		J	
5h 5.id	160y	TTC/17 946 964	100	72.4.1	7.0d	2 24h 7 747m	195m 6.5h 8 83- 8 14.17	3.04m 8-3 2			100			
2 4 122,089 2 9 2 08	y 154	10 94 8 85.	σ,21	.879.299.966. 0868-139	0256,0499	25,89,726,.81 81 .89,081	7 35, 39, 7 33, 03, 49, 07, 7 16, 51	756,6/1, 215, 169,061,44			102			
<u>Ev2 4</u>	E.07	E120 E 95	158.9254	675×61 21,83	E 16	E-1,2.8	R4 E1.7				-			
Gd 155 3-	G0 156	Gd1574-	Gd 158	Gd 159 V- 146ms 18.0h	Gd 160	Gd161 <sup>(5/)</sup>	Gd 162				ļ			
a 5 1 10 <sup>4</sup>	σ, 8	a.2.50.44	σ, 2.4	11. 70 9 95,89	σ <sub>¥.</sub> 17	18 1.56 y 361, 315, 102,	y.41,.43							
	IRE 00200	186 0240	1770242	y 363,0580 07951,85	(50.027)	0 y 10 3								
Eu154	Eu 155 54	Eu 156	Eu 157	Eu 158	Eu 159	Eu 160		· · · · · · · · · · · · · · · · · · ·	L.,	l	1			
7 8y 8**** 25.87 (85	5 Oy 8 - 5 - 244	152d	15.2h	45.9m	18.1m 8-257:9010-	~2.5m			100					
123.274,723 1304_058_159#,	0865 /* 014 46	• 0690,199 - 519 - 21	-41,0650,16- .97	y.,380, 95, 89,	175 x067,67,092,				100					
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5m 155* 46.8h	22.71	22.3m	5m156 94h	5m157 05m										
8 70,65,80, 103,0697,	α.5	8 1 14. 7 1044, 246, 141	81 43, 70 9.087 20, 04-	β- 1 x .57										
083-636 E 801	153,9223	026-130 E165	.29 E 72											
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Тт I65 <sup>1/+</sup> 9µ = 30 h 040,03) = 8 30 80µ = 1004,243 9 016 - 443 9 107 = 16 - 37	Tm166 <sup>2+</sup> 7715 06: 93/2 268 304	Tm167 <sup>1/+</sup> 9.3d 7.208, 057 7.5 - 532 E1,16	Tm168 <sup>(3,1)</sup> 93.1d v.198.816,448, 184,0798-149 5.172	Tm1691/4 100 σ <sub>γ</sub> (10+103) 168.9342	Tm170 4 μ4 129d 144, β-967, 88 058, -7 0843 6, 7 γ95 Ε 967 Ε*5	Tm171 1/4 2.5µs   92 y y 308, 8 098, 112, 0.67 0051; y 0.67 y 4.5 E.098	Tm172 63.6h 81.0.188,	Tm173"4 8.2h 8-86,.90,	5.5 m β <sup>−</sup> 12.7 y 3670, 9920, 2730,- € 3.0	Tm175 <sup>(14</sup> 16 m β <sup>-9</sup> ,15,19, y 36,94,042- 1.51	Tm176 1.4m 8 <sup>-2</sup> 0,115,3.05 7.19,105,87,1.8, (.096~.389) E4.1	
Er 164 1.56 0,13	Er 165 *-	Er 166 33.41 0;(12+17) 165.9303	17-Er 16774 2.35 22.94 17 208 e g65 16 166.9321	Er 168 27.07 σ <sub>2</sub> 191 167.9324	Er 169:/- 9.3d #:34.33 7.0084 e* 1.340	Er 170 i4.88 σ <sub>γ</sub> 5.7 i69.9356	Er 171 5/- 7.5h 8-106,58-149 3083,2959 .1116,012-140 F1,490	Er 172 49h 8 <sup>-</sup> 28,36,9 9,05,41,61,0	Er 173 12.0m 8°2.3,1.8 7.20,40,18, 037-0.52			108
140 <b>163</b> 172 - 219 2	Ho 164	Ho 165 <sup>7-</sup> 100 0y (3.2+62) 164.9304	*H01660 (2x103y26.8h 8~06, 81.85, 0806, 175 (184,8h p.0806, 712, 1380 712, 1380 21ms £185	Ho 16717/1 3.1 h 8.32 97, 61 9.346, 321, 079, 2080, 10 74- 745 E 0.9.7	Ho 168 3.0m 8 2.0 8 5	Ho 169 <sup>721</sup> 4.7 m # (20,195, - y.78, 85, 06- .92 5.21	Ho 170 44s 8= 3.3,			106		
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- 24 -

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<del>69</del>	168.9 <b>34</b>	58s  ຂ5%	2.985 55 9504 9496						30m € y.046373 E35	79m 22m € €+2.3, 24 3.8,9 >	1.8h €, β+11,.4 γ104,242,.022 1.80 E2,27	I.9m β+294,,€ γ.091,.21-238 ε 3.96	9µs 30h 7.060,031 €,8*30 80µs 016-143 17.069 e- 8+1.7 e- E2.1 E1.57
	9,37 Tm	Tm153	Tml54				 	·····	Tml6I	Tm162	Tml63 <sup>14</sup>	E 2 7 Tm164 <sup>(1+1</sup>	ε- ε-3 Tml65 <sup>1/+</sup>
70	173.04	0.395	16s a 52i						~ 23m	IIm	77m 6 8†29	10m #*158,,4 y 080,069,	57h € y 082
	σ, 74	Yh154	Yhi55		·		 		YhI62	Yb163	YhI64	Yhi65	056-40 E30 Yhl66
71	Lu 174.97	0.07s	0.235 ~0.55										55 m €,8 15,11, 1 y 030, 239,
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I I			n Anglaith Nan	And the second sec	Frica 1777	Re 178	Re 17954 20m	Re 180 2.45m 6.8118 7.512, 104. 076-825	Re 8 (5/4 19 h , 1.366), .361 639, .020	Re 182 12.7h 64h 48+17.55 4 140.0647 y 020 032-7.05 144	25 R e 1835 1.0ms   70d 1.1:34 € 7.1:30 r.046, 62.053 041:40	165d 38d 11 083 4 76 110,216 74 110,216 79 033 110,216	Rel85* 37.07	Ŧ
<b>Arroy 1</b>		W173 16 m	W174 29m	1	W176	W 177	W 178 2150 Filet ag. Bob 1, 093, 20147	(138 (11) N 179(7) (11) N 179(7) (12) N 179(	18 W 180 5.4ms 0.4 11 391 0,~ K 9.450 15. 234 10.4	(E2.86 <sup>M</sup> →W18  <sup>9</sup> /ι+ 14.6μs  130( 11.366,006Γ 252,006Γ 7113 133,13	W 182 26.41 α <sub>y</sub> 20.7	1.37 ε~16 10 <b>W 183</b> / 5.2 s 14.4 17.102 210 7.105 05.5	184.9631 W184 30.64 cc(2mb + 1.81	
	÷	To 172 44m	Ta:173	16174+ 2	TO (15)**	1 Tal760 816 9 1902,	F 09 Tal772 56 6h	2:9, 1. Tai78 9.3m 2.2h 6.8' 69, 6.8 60 y 332 7.0932, (089) 20-147 (089)	e (79 <b>.947</b> (94 <b>)TQ</b> [79(74 14µsl ~600d 7.031 е пот	E 19 1 <sup>(4)</sup> Ta 180 <sup>(6)</sup> 8 13h 0.0123 7 0930 8 -62, 51.6x10 <sup>13</sup> 71 6, 8 <sup>-</sup>	181,9483 1/+ <b>Ta 81</b> 7/- 8,2µ3 99,98 171,53 a. (01+2 7,482, 180,948 346,156,-	046. 182.9603 1841 To 182.9 15.9m 115d 11.165, 356,8 516 7,172, 47, 2543, 256, 1319/1001 276, 1319/1001 276, 1319/1001	183.9510 T0183 <sup>7</sup> / <sub>4</sub> 5.0d β=.62. γ2461.1079. 1614.2443. 0410-407	- *
Hf169 1.5h 81 2 4.5 1 91, 19	Hf170 12h 1345	Hf171	Hf 172	HE 73 KAN A. Maria	<b>1:11.724</b> 0.17 2:0 - 0 <sup>1</sup> 4:0 - 0 <sup>1</sup>	$\begin{array}{c} 0 & \text{Hf} 17507 \\ 154 \mu s & 200 \\ 156 & 143099 \\ 4 & 432 \end{array}$	Hf 176	L 9 234 Af 1777 L's 18.5 3941 246 374 3841 246 374 3841	E 115 Hf 178 >10y 27,4 126. 4152434 574. 177943 556. 173, 5669 458 htt case 155	17 102 179 9475 17 Hf 1799 18.65 13.75 11.161, ct.33+ 375 7 217	Hf 180 55h 35.24 11.0575, 07 12.2 501 179.947 332.	- 42.4d -	Hf 182 9 x 10 <sup>6</sup> y <sup>β-~4</sup> y.270,	-
Lu 168 71m 12/0h? 8190 cy 087 087	07-124169 774 2 7m; 1.53 11 029148 - 19 4 17 087,664 - 087,664 - 071	4-1927 0.75 0.66 0.48,68 9 0.65 084,99 0.65 084,99	рана раз раз раз раз раз раз раз ра	ante provincia de la composición de la composi Composición de la composición de la c	17.39404 1949 17312 47.46 5.844 17.24 5.844 17.24 5.75,079	Lu174		176.9434 176.77 3.59hi 2.59 8.123.2.6 x10 <sup>6</sup> y 7.86, 5 31, 202, y.186, 5 31, 202, y	236.21148.091 237 LUI7777 151.00 (6.71c 6.10167449 17.16 (7.206.0 17.16 (7.206.0)	$\begin{array}{c} & (78,546)\\ (7^{-}) \left\lfloor 0 \\ g^{20m} \\ g^{-} (2 \times 8) \\ g^$	E114 Lu179 <sup>(74</sup> 4.6h β=135,108 γ-217	Lu 180 2.5m β <sup>-3.3</sup>	E~.5	
×46 Yb167(5/1- 17.5m 27.165 26.1130, 26.1132	42.25 Yb168 0.135 σ, 3.200	+ 14 • Yb I69 • 46% 11 тоте • сов. 198 • с7, 02+ 108	Y5170 3 03 7, **	Yb17110 1431	Yb172 21.82	Yb173 35,2,3 16.13	174 9406 19 b 174 0.80ms 31.84 10 c 40 g (46 - 0.40 m 19)	17401 L102 11/175174 67msi 419d 11513;β467, 1396,283, 114,	(12.73) (1.55) (1.55) (1.55) (1.2.73) (1.55) (1.2.73) (1.	D889) F21 VYD 7794 641s 19h 1723 8 140, 71040 76, 176, 198, 139	E135	£3,3		1
196 Tm166 2* 771h 7.8*19312 7.081,184,07 2.68	167.934 Tm167 // 9.3d 7.208, 057 25 532	Тті 168 198 ғылаан 194, 179 – 43	Tm169 100 «2002-1031	170 9364 Tm170 3 545 - 104 1 144 - 2 16 - 169 1 144 - 2 16 - 169 1 144 - 2 16 - 169 1 144 - 2 16 - 169	сті азена Пом. 1711 2. п. н. 1925 4. п. н. 1925 1061, соны 1	<u>172 9581</u> 6 Л (л) 8 - в.: 48, 	825 7.400.47	E 467 Tm174 5 5m B - p 7 - 56 76 - 9926 - 2730	16.m 16.m 8 9,15,19, 36,94,042	1/40 Tm176 1,4m 872 0,115,3.05 v19/05,971.8, (096 - 389)			110	]

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410.253.4922         75.71.074         77.746           516.064         71.074         71.774           52.9         10.1         98           411.105         10.1         98           111.105         10.1         10.1           111.105         10.1         10.1           111.105         10.1         10.1	41(2)     23(2)     23(2)     23(2)       94(2)     41(2)     23(2)     23(2)       94(2)     10(2)     53(2)       11(193)     11(2)     53(2)       11(193)     11(2)     53(2)       11(193)     11(2)     53(2)       11(193)     11(2)     53(2)       11(193)     11(2)     11(2) <td< td=""><td>/</td><td>16m 75d 7.75 (422) 7.75 (422) 7.7</td><td>28.41 07.36 185.9544 TO 185<sup>0041</sup> 4.9m</td><td>23.95 s: s3, 32, 34, 54, 54, 54, 54, 54, 54, 54, 54, 54, 5</td><td>69d 8:349 7:290.227 064 5:349</td><td>11.5 m e 2.0,2.5 ; 258,417, 094-095 E2,5</td><td></td><td></td><td>118</td><td>120</td></td<>	/	16m 75d 7.75 (422) 7.75 (422) 7.7	28.41 07.36 185.9544 TO 185 <sup>0041</sup> 4.9m	23.95 s: s3, 32, 34, 54, 54, 54, 54, 54, 54, 54, 54, 54, 5	69d 8:349 7:290.227 064 5:349	11.5 m e 2.0,2.5 ; 258,417, 094-095 E2,5			118	120
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Os188 V Os189 Visc Os190 V Os19 PC 13.3 5.3 h 16 i 14m + 264 J30h 155d 1.3.6 i 15m - 4 i 15m -

161 67

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Tm166 2+ Tm167 + 771h 94d 0.061,07 - 208,057 268 07 - 208,057 268 - 532 404	100 163 9700 Tm168" Tm1691 100 104 107 444 104 1070 443 104 1070 443 104 1070 443 104 1070 443 104 1070 443 106 3342	170 4164 Tm 170 1744 History (171 1744 History (	172 9590 173 9590 170 172 - Trn 173'44 6 - 3 (m 8 - 24) 170 - 44- 170 -	$\begin{array}{c} 1.4, & 3.4, \\ \underline{E} 467, & 3.4, \\ \hline Tm174, & Tm175 \\ 5.5m, & 16m, \\ 5.2, 7, & 8.9, \\ 5.4, 5.4, \\ 5.5m, & 3.4, \\ 5.5m, & 5.4, \\ 5.5m, & 5.4, \\ 5.5m, & 5.4, \\ 5.5m, & 5.5m, \\ 5.5m, & 5.5$	e 194,333 E140 Tm1/76 14m 9720,1E,305 979,05,97,08, (096-389) E41	
98	100	102	104	106	108	

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77	<b>78</b> Ir 192.2	195.09 مران Ir171 LOs	a 629 Ir 172 25 a tota	a 693 Ir173 35 a 567	a 5.95 Ir 174 45 a 5.48	a 5.73 Ir175 a5.39	a 5 52 Ir 176 Bs	a 5 44 Ir177 215 a 544	a 5 15	g *, .4	a 5 02	e a 4 R4 Ir 181 Short	Са473 Ir 182 15m с в т	17, 183 58m 62, 44 58m 62, 844,0 58m 6
76	n <sub>o</sub> 426 Os 190.2 g <sub>a</sub> 15							Osl76 3.6m 2.76.0.090.857		Os178 5m	05179 8m	Os180 24m (, 190	y (3, 28, - Os [8] 2,8m, 105m €,2 <sup>21</sup> (5) 105, -y 239, 8, -y - 108, - 107	> 24, Os 182 <im &gt;</im 
	a	94		96	1	98	l	100	1	102	L	104		106

	108		110		112		114		116		118	
7	103.9507	E 98	(85.9530	185.9008	187.9561	108.9583	<sup>167</sup> 168.9586	E 310	191.9615	Gy~1500 EU32	.097	£2.0
171 <b>7.38</b> 2, 0	ey, 3.0 x 10 <sup>8</sup>	y 65, 88, 87, .07272		(10,57).		σ <sub>γ</sub> (20mb +7)	y 503, 6-7, 361,	γ 042D, 1294,	σ <sub>y</sub> 2.0	y 46i, 139, 559, 28i, 073 - 874	y .043,	βζ
9h   14h	0.018	946	1.59	231µs 1 <b>.64</b>	13.3	59h <b>16.</b>	9.9m 26.4	13.0h 15.5d	41.0	31h 8.03.06	6.0y	6.5m
Os1834/#	Os 184	Os 185#	Os 186	Os 1874	Os 188	* Os 189*	<sup>10</sup> 0s190	<sup>17</sup> Os 191 <sup>64</sup>	Os 192	Os 193	Os 194	Os 19
	_	1495,071 E383 2.75	£.~L6	E284	NOI 11	4791.2h 11.026	082 190.9606	17.208 489	192.9630	γ.33,29,59·21 12,25		-1.564 E3
164, 120, <del>391</del> 4, 3	y.024 - 667	7 137, 7137, 297	965 (j) 665 (1) (1)	y 1550, 633, 478, 31, 2, 2	નાવેલ્ટ અન્વસ્	11/149	y 129, 7 +436)	CEO5H (36) 139	7,000+10 <b>7,11×10</b> * 2,121	084 192	120-800 1296	7 355, 7 779, 333 57
9*	6	8193 1.81 94.	L-D 🗶	#,A*166.12,	17.07Z, 17 1940 - 2020	\$ 187, SRI 19 187, O'SH	11 042 0 (4+ 52	101 4m s aca	11.080	Sims 182.24	-685, 18 2.(1.3)	$\beta^{-3.2}$ $\beta^{-}$

			11 191	M1 192	MIT1 193	TI 194	₩TI195	T11962	PTI 197	7+TI 1982	9/11199	17+1T12002-	
			۰0m د	1165 1 1065 16 - 17 16 - 18 -	2.181 - 2.081  1.14.095(x  x.365 - 14.24	202.0FN - 34/B €	13.05 1.1711 11.099 €,8*~18 1×383 ∞037	€ 7084 € 7084 7 426	17 222 € 17 385 y 152,	€, y.048, €, 8*24 2.1,	11.383, € 	1T0.21 €,β+ γ0.538 1.06	
		1			25, 26 27 - 33	a 5.82 E~5 4	226 BF	11 120 E4.6 y234, 241, 275	.018 - 1.0 E.2.2	11.261 y.412 y.283 195- E35 2.1	- 367, 35, 247, 156 - 72 Et. 1, 035-49	y 368,121, 116-2.01 E2.454	
Hg 187	Hg 188	Hg 189	Hg 190	Hg 🕀	Hg 192	11Hg193*	Hg 194	WHg195	Hg 196	13/+ Hq1971/	Hg 198	12+Hg1991/-	
3m •.β	5./m €.ø•	87m	20 m	568	6* 	ilih 4h ∈,β*πζ €	0.40s ~ 1 3y	11 123 E	0.146 g(120+3.0x10	23.8h 64.1h 11.65, €	$\sigma_{\gamma}^{(10,02)}$	43m 16.84	
a 5.142	7014 114, 193	y 17, 24, 32, 50	y 029, 145.	255 274	1 - 2 AN 19 1 1951 4 M A - 1 M	r 251, 187, 11102, 76,	134	053 9,057 6,16,56 6,16,56	195.0650	279, .191,	197 96676	7.106 0,~2000	
Au 186	Au 187	Au 188	Au 189	Au 190'	Au 1913	Au 192 -	W Au193	Au 194	"Au 195**	2 Au 1962	-Au 197	Au 1982-	
2m €,β⁺	Q14s 8m	8 m 4	4.6m 30m	42m	3.2h	4.7h 6.8*249.219	3.9s 17.6h 11.032. 4	39.5h ∉ 8*149	30.5s 184d	9.7h   6.18d [1.175   ε,β+ 1.148   ± 356 33	7.5s 100 TT.130 c.96.8	2.696d #=.96,.29,1.37	
y 16, 22, 30, 41	noy	7 23.34, 61		y 751, 50 3.44	v 091, 048, 16, 13 - 03 - 2,17	y 317 296 05 3:04	290 y 86,112 y 258,- U.5 .050	y 329,10 2 41	, 26) 031-211 200,80	188, <i>B</i> 26 y 426	y.2/9,	y.4118,.675, o, 2.6 x 10 <sup>4</sup>	/
Pt 185	Pt 186	Pt 187	Pt 188	Pt 189	P1190	Pt 1911	Pt 192	1231 11/1971936/1	Pt 194	134Pt 1954	Pt 196	13/1Pt197 -	
Lih	2.3h	2.5h	10.26	116	4544 00127	107µs   3.0d	9545 0.78	4 3d < 500y	32.9	4.1d 33.8	25.3	80m 20.0h 17 347 β 67,48;	
y 055, 63,156	6 0.4.23	B', 4 y 10, 106, 180, 192, 2,11	y 195, 188, 1955, 1351-478	€ ↓ 725, 16 5, ⇔*>+ 01	7310 y 03.18	ст (94) 97),48) 97),48)	ayı.	y.013 noy	a;(.09+1.1)	7 099. 0.27	(¢(1.05+.9)	7.0502 7.0779 B 191, 269	
	.68, i 40,	477 1.00	a 192 1.51		169.9600	L	191.9612	E 05	193.9627	194.9648	195.9650	279 E.75	
Ir 184	Ir 185	Ir 186	29ms (11.2h	Ir188°)	11/1 Ir 189 (3) 12ms   13.3d	11 17 190 (4) 3.1h   12d	<sup>11</sup> Ir 191 <sup>34</sup>	~650y 74.3d	<sup>1</sup> Ir 193 <sup>34</sup>	1714 Irl94	4.lh 2.8h	Ir 196	
€, <b>8</b> * × 264. 12059	¢ v.024 · 667	8 <sup>+</sup> 193 * 8 <sup>+</sup> 194 137	1.0.0	# 8*166.12 + 1550633.478.	17.072, 14 258 - 4070,	9,8720 1,187,961,9197,091	(1 042 g (4+52 y 129) 7 +436)	11:361 07:67:54 11:31:4m y ws. 468 17:054 136 138	11.080 0,105+0,11×10 <sup>2</sup>	3ims 8 7.112, 2.24	7.685, 8°2.0.3 433,319, 7.0987,	β <sup>-</sup> 3.2 β <sup>-</sup> 1.16 γ.355, γ.394,	
4.3		4 95, 071 E 4 95, 071	ing` D €∼56	31 2 2 E 2 84	301 225	17-01.2h	047, 082 190,9606	5 / #* y 201 489	(2.62) ( 192.9630	y.33,29.59-211	120-000 1230	-1.564 E3.4	
"Os183"	Os 184	Os 185#	Os 186	Os 1874	Os 188	° Os 189*	<sup>10</sup> Os190	<sup>17</sup> Os 19/84	Os 192	Os_193	Os 194	Os 195	
9.9n   14h 4 11  71   14a		940 €	r28	2.39μ <b>*   1.64</b> [110.157]	15.5	59h <b>16.</b> 1.0309	9.9m 26.4 11.039 ac (10+3)	15.01 15.5d	41.0	- 31h -β (13,106, - 461 139 559	6.0y β=.097,.054,	6.5m ∦°2	
71.10 1030, 115, 145 1030, 1.44		.07272				+2)	7 305. 6:7,361, 87	1294,	y	281,073-874	y.043,17		
067	103.9607	E 98	185.9530	185.9338	187.9561	188.9583	198.9598	E 310	191,9615	γ~1500 EU32	E.097	E2.0	

![](_page_201_Figure_3.jpeg)

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							86	Rn	i	Rn200? 3s ¤ 6.77	Rn201 3s a 672	Rn202	Rn203 28s   45s 2655   4650	Rn 204 75s a 6 42
			85	At	At 194 Short	At 195 Short	At 196 0.3s	At 197 0.4s 66.96	At 198 1.5s 5s 16.85 66.75	At 199 7s	At 200 4.3s   42s 0.654   6641, 0.46	At 201 1.5m 4634	At 202 2.6m2 3.0m	20399 At 203 7.4m € α 6 09
		84	Po	Po 192 0.5s a 6 58	Po 193 Short a 5 98	Po 194 0.6s 0.65	Po 195 2.0s 5s a 6 70 a 6.61	Po 196 ~ 5.5s a 6 52	Po 197 265 565 16.58 4.6.28	Po 198 1.75m a 6 18	Po 199 4.2m   5.2m å4.06   å5.95	Po 200 11.5 m 6 5 Ho	<u>іс чел</u> 9.0m : 15.4m й б.7н : 15.6н	En 68 Po 202 44m 6 a 5 58 F = 10
	83	Bi 208.980 <sub>06</sub> 34mb	Bi 190 ? ***	Bi 191 ? 145 a 6.9 a 6.31	Bil92 ~ 40s 40s	Bi 193 3.2s ~70s a 5.5 a 5.89	Bi 194 85s 956	Bi 195 ~60s   2.5m a 610   a 5.4	Bil96? 7.8m	Bi1972 1.7m a 6 2	Bi 198 11.9m	Bi 199 9/ 24.7m 7 5 50	Bi200 35m 6 1/04,462 6 5	Bi 2019- 53m 111m 62525 y 6290 E 42
	82	Pb 207.19 <sub>6.18</sub>						Pb 194	Pb 195 17m \$.0990, 383	Pb 196 37m 5/92, 240, 253, - E2 8	42m 2 42m 2 44m 2	Pb 198 2.4h € 7.75.,190, 031.865 E~1.8	12.3m i.5h 12.3m i.5h 11.424 e.8728 11.424 e.8728 11.424 11.57 11.51 12.57 11.51 12.57 11.51 12.57 11.51 12.57 13.57 14.57 15.57 14.57 15.57 14.	Pb 200 21h 7 (48, 142, 235, 258, 033 (45) 8 (19)
81	ΤΙ 204.37 σ <sub>6</sub> 37					T   9  <sup>IOm</sup> •	P¶[   92 ≥)   m    0m € y   ,42	(9/1)TI 193(1/) 2.1m 23m 11<.025 y.365 y.24, 25.26, 2733	TI 94 32.8m 34m € .097 ¥ 43 µ 58? E~5.4	8/ 1 <b>T</b>   195 <sup>1/4</sup> 3.65   1.17h 11.099 (€, β <sup>+</sup> ~1.8 7.383   7.037 226 88 €3.0	121) <b>T1196</b> 0 141h 18h *084 <b>*</b> 10120 E4 6 1034 241 275	9/T1 1971/4 0.54s 2.83h 17222 - E y 385 1y.152, 018 - 101 E2.2	7:T1 1982- 187h 5h 6. y 048, 6. 8+24, 21, 17.261 y.412, y 283, 1952,8	9 TI 994 28.7ms 7.4h 11.383, 455, 029 7.455, 367, 35, 208, 727 247,158 ELL 035-49
		106		108		110		112		114		116		118

 Rn 21744
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Rn225

Rn226

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									90	Th 232.038 g <sup>74</sup>	Th2 3 .15s ¤ 769	Th214 .13s ¤ 768	Th215 1.2s 0.7.39,752, 733
						89	Ac	Ac209 0 ls	Ac2IO ~0 35s @ 7 46	Ac211 ~ 0.25s a 748	Ac2 2 ~0.93s a 7.38	Ac213 0.8s a 7.36	Ac2l4 8.2s a7.21,708, 7.00
				88	Ra	Ra206 0.4s @727	Ra207 1.3s a 7 13	Ra208 1.2 s a 7 (3	R0209 47s	Ra210 385 0702	Ra211 155 211 001	213.01 Rd2l2 13s a 6 87 212 000	214.01 Ra 213 2.7m a 662.673, 652 213 000
		87	Fr	Fr203 0.7s	Fr 204 2 2s = 3.3s 27.15 = 9.597	Fr 205 3.7s 9 6 92	Fr 206 15 7s a 6 79	Fr 207 15s 9677	Fr 208 38s 9665	Fr 209 54s a 6 65	Fr 210 3.0m a 6 57	Fr 211 308m a 6 53	Fr 212 19m 46 26.6 38.6 41 6 34.4 E~57 2.0 0
86	Rn	Rn200? 46.77	Rn201 35 4677	Rn202 11s 26.64	Rn2O3 28s 45s 76.5° 76.50	Rn 204 755 a 6.42 203.99	206.00 Rn 205 2.8m a 6 26 204.99	20730 Rn 206 6.3 m a 6.26 ¢ 205.93	208-30 Rn 207 ∐m € a 6 13 E ~ 4 8	<b>Rn</b> 208 22m € α 615 €~30	Rn 209 30m ε α 6.04 ε~4 0	Rn 210 2.4h a 6 04 6 209.9895	Rn 21   1.1-7 15h * 032+180 a576,585,562 7.069, 17, 23 E2,89
	112	114		116		118		120		122		124	

Pa 237 39m 2 30,1 35,---7,46,92,09-1,4 E 2.30

Pa 235

Pa 234 UX<sub>2</sub> j UZ

Pa 233

Pa236 <sup>12m</sup> 8-3.3

			7340y 7340y 7340y 7494 491 7, 0 7, 0	Th230 Io 78xi6*y 78xi6*y 7.04	Th23104 2552h 268 268 268 268	Th 100 n L41 x 10 y 4 01 3 35 5 7 4 01 4 100 5 7 4 01 4 100 5 7 4 01 4 100	Th233 22.2 m β <sup>+</sup> 24. γ 229 - 85 η 1450 - σ <sub>1</sub> 15 Εί 245	Th234 UX, 24 IOd 3 19,00 7 05990 1.30- 9,016 094 1,27 77 020	Th235 < 3m				150
			Ac228 3 MSTh2 613h A 24, 24, 24, A 34, 049, 40, A 34, 44, 44, 44, 44, 44, 44, 44, 44, 44,	Ac229	Ac230	Ac231 15m 8 20 8 20 8 20 8 20 8 20 8 20 8 20 8 20						148	
			Ro227 412m 9 - 31 7 29, 56, 027	Ro228 MsThj 575y Msthj 575y Mstate Mstate Futos	Ra229	Ra230							
			Fr226					144		146	-		
			Rn225 4.5m	Rn226									
			l	140	1	142	]						
					~								
						Pa	Pa224	Pa 225	Pa 226	Pa227 <sup>(5/-</sup> 38.3m	Pa 228(3) 26h	Pa229(5/-1	
			t	THOOD	91	71,000	1 T-007	a 7 25 225 03	a 6.86,6.82, ~ 6 E 2 78 226.028	a 6.47,642, y.065, E 1.00 227.0288	5058-1.89 a6.08,5.71-6.14 y.1335 E 2 10 228.0310	5.0424 a5.58.5.67, 5.32-5.74 y.026-071 E.35 ThOOO	/
1 h216 25ms a792	10217 5 5ms 11925			10220	1.7ms 1.7ms 13= 6 4 4*	1 N222   ~ 2 8ms   1 98	065 a 73	111224 1.1s a717,699. y 77,	111∠∠⊖ <sup>194</sup> 8m a648,631-680 y 322, 362, 246 €	31m 31m a 6.33,622, y 10, 242,131,	RdAc 18.72d α 6 0375.958 5 755 7 236,006 - 443 σ <sub>1</sub> ~ <u>450</u>	RdTh 1.913y a542,534, y.084, 216, 13, 17 $\sigma_y \sim 123, \sigma_f < .3$	
Ac2!5 017s 9.7*	AC216 0.38ms -0.38ms 9.91 - 2.9 (13, 9.07 (820,828		Ac218 Short	Ac2l9 Short	Ac220 24ms 0.4:00	Ac 221 0.055 0.754, 7.43	Ac 222 5s a 7.00,6.96	Ac 223 2 2m ac 648,6.659,	Aç224 2.9h 2.9h 2.9h 2.6.132 46.044,6203, 6.139, 5.149,5.20,5.39,5.5,5.5,5.5,5.5,5.5,5.5,5.5,5.5,5.5,5.	AC 225/3/- 10.0d 5.82,5.79,5.73; 7.037-,529	Ac226 29h # 88.11 70721-230 5.68-26 45.44?	Ac 21.772y B 044 G495,4.94, 7,005 19 E.044 7,790 E.044	
Ra214 265 2714	Ra215 1.56ms an /0, / MK h	Ra216	Ra217 <03ms a 900	Ra2l8 Chort	Ra 219 IOms 2768 794	221 0:57 Ra 220 ~ 23ms a 746,690 ; 465	222.0-78 Ra221 29s 6.665,- 7089,152,176,- 200-000	223 0191 Ra2222 385 26.56, 7.325,	AcX 11.43d a 5.714,5-605, y 270,031-580 g 130, g 1 - 32,005	<b>Ra224</b> <b>ThX</b> 3.64c a5684.5447 y.241, 290-55 oy 12	$\begin{array}{c} \mathbf{Ro}_{225}^{1.12} \\ \mathbf{Ro}_{225}^{1.5/2} \\ \mathbf{I4.8d} \\ \mathbf{\beta}_{7040}^{-32} \\ \mathbf{y}_{040}^{-32} \\ \mathbf{r}_{10}^{-32} \end{array}$	Ro 1602 y 4 781,4 598, y 186, 260 - 610 g 20, g, < 1mb	/
Fr 213 347s	Fr2l4 34ms' 50ms 448' 0442 848 840	Fr215	Fr216 Short	Fr 217 Short	$\frac{1}{219500}$ Fr 218 $\frac{1}{3} \times 50\%$	Fr 219 21 ms 7 1 40.	Er 220 28s a 6 69,61.4	Fr 221 4.8m a 6 34,612, y 215 063 4 2 108	Fr 222 15m 8 <sup>-</sup>	Fr223 <sup>3,9</sup> AcK 22m β 15 γ 150,080,061-96 μ 5,54 1 1 49	Fr 224 2.7m	Fr225 3.9m	
Rn 212 25m 25.0	Rn213	Rn2l4 Short	206-05 Rn 215 ~ 1μs α 8.67	Rn 216 05ms 05ms	Rn 217	Rn 218 55ms a 1.1 × 5a y 1	Rn 219 An 3.96s	2210:42 Rn 220 761 55.65 66 286.5 747 7 542	Rn 221 25m 25m	Rn 222 Rn 3.824c	Rn 223	Rn 224 <sup>19h</sup> #-	
<u>, 9967</u>	denist	214 11	214 999	2.6 (9.9.3	2.7.00 <b>39</b>	2-8 JU56	2 (2 - 2 - 1) - 2 - 2 - 2 - 2 - 2 - 2 - 2 - 2 - 2 - 2	220 01:4	£~10	222.0175			

Po 2313- Po 232

Pg230

#### 126 128 130 132 134 136 138

227-App-C

![](_page_206_Figure_1.jpeg)

					105					260? > 10ms a 9.7	261? 0.1-35 a 9.4		
					104		-	257 ~4.5s α 9.00,8 95,8.78, 8.70	2582 Hms	259 ~3s a8 77,8.86	260? 0.3s SF	261? ~Im ¤8.2-8.3	
			103	Lr				Lr 256 ~ 35s a 8.4,~	Lr 257 a 8.5 8.6	Lr258 or 259 <sup>8s</sup>			
		102	No	No 251 0.85 a 8.60,8.68	No 252 2.4s 3F	No253 L6m a 8.01	No 254 56s sF	No 255 3.0m a 8 06	No 256 3.2 s a 8 43 SF	No 257 235 a 8.23, 8.27			
	101	Md				Md252 8m	239.091	233.09	Md 255 28m a 7.34 E~.85 255.0906	Md 256 1.3h a 718	Md 257 ~ 5 h a 708 SF	Md 258 54 d a 6.73,6.79	
Fm245	Fm246 1.35 a 8.24 SF	Fm247 9s   35s a818 a 7.87, 7 9 3	Fm 248 .61m 0 787, 783 35 248.0771	Fm 249 ~26m a 7.53 249.079	Fm 250 30m a 743	Fm 251 7h 4 6.9 7.41 End 2	Fm 252 23h a 704, 7.00 SF 252 0826	Fm 253 2.6 d 6 695,6 68, 7.145, 272	Fm 254 3.24h a 720,716,7.06 y.041, 098,.15 SF 254,0868	Fm 255 <sup>77</sup> 20.1 h a 702.6.97.6 41- 7.08(.059, 5F ay 26 255.09	Fm 256 2.63h sf ¤ 6.9	Fm 257(9/+) 80d a 6.52,670, 6.44 7.062,18, .24, S F	Fm 258 < 0.2 s
	Es 245	Es 246 7.7 m	Es247 5.0m a 7.33 Ev 2.3	Es 248 25m 4 5 8 8	Es249 2h a 6 77 E 1 4! 249.0763	Es 250	Es 251 1.5d ¢ a 6.48 Erv 6 251.0799	Es252(7+) ~ 140d a 6 64,5.9- 6.6 y.40,07-57 252.0828	Es 253//+ 20.5d a 664,5.73-6.63 y 04(8, 0088-90) SF 0,560 or (180 + 14) 253.0847	2 - Es 254/1+ 39.3h 276d 6 48,113 664, 6 48,113 664, 6 49,114 605 6 49,114 605 7 605 6 49,114 605 7 605 6 49,114 605 7 605 6 49,114 605 7 605	Es 255 39d a 6.31,6.27,6.22 SF o <sub>y</sub> ~ 40	Es256 <sup>22m</sup> β <sup>-</sup>	Es257 <20h β <sup>-</sup>
Cf243 i'm ¢ a 7.06, 7.17	Cf 244 20m a 7 21 244.0670	Cf 245 44m a 214	Cf 246 36h a 6 76,6 72,- 7 042, 096, 146 SF 246 0688	Cf 247 2.5h 2.255 42,46 Ft 9	Cf 248 350d a 6 27, - SF 248,0723	Cf 249 <sup>9/-</sup> 352 y a5.81,590,593, 6.70,- 7.366,353,253,- 51 g,~270, g,650 360,0745	Cf 250 13.1y a6.03,5.99 y 043 SF cy ~ 1500 0 < 350 250,0764	Cf 2511/4 900 y 95.67,5 84,6.01,	Cf 252 2.65y a 6.12,6.08, y.043, 100, SF or 20 252.08	Cf 253 17.6d $\beta^{27}$ a 5.98, 5.92 $\sigma_{y} \sim 165$ E.27	Cf 254 60d SF 0 5.83, 5.79 0 y < 2		
	Bk 243 46h 7.75, 95, 84 676 54,618 676 7187, 04:55 £149	Bk 244 44h 7216, 89, 145 1.51 06 67, 6 62 Ev 2 2 244 0 7	Bk 245(3/-) 4 98d y 253, 38(, 385 3 289,615,6 36, y 207, 87, 166 E 84	Bk 245 2 18d * 80,108,734-	Bk 247 14 x 10 <sup>3</sup> y a 5.52,568,531 y 084,27 247,0703	Bk 248 <sup>(c)</sup> >9y 16h a a a 55 8 (sr 65 (sr 65)	Bk 249 <sup>2/+</sup> 3msi 311d 3 <sup>1/-25</sup> 1a 5.41 1y 3.47 307	<sup>211</sup> Bk 250 <sup>[2]</sup> 2[ms] 3.22h γ.043 β 73,176 41 γ.99,103, 29μs /τ <sub>7</sub> ~1000 γ.036 [ε] 76	Bk 251 57m β~ 5,~10 γ 037, 094, 140, 184 Fo.1.4				
Cm24(11/4) 35d 7 47, 60 a594,573-608 7 140 F 77	Cm242 163d a 611601 2,044,000 94 2,0544,000 94	Cm243** 22, 24, 27 24, 24, 27 24, 24, 24, 27 24, 24, 24, 26 7, 24, 26, 4 7, 24, 26, 4 7, 24, 26, 4 7, 24, 26, 4 7, 24, 27 7, 24, 27	Cm244 34m3 - 84 y 175 - 95 - 85 y 175 - 95 - 85 y 175 - 95 - 95 - 95 y 175 - 95 - 95 - 95 - 95 - 95 - 95 - 95 -	Cm245 <sup>24</sup> 5 3 x 10 <sup>3</sup> y 5 3 x 5 5 5 7 1 <sup>3</sup> 5 5 5 7 1 <sup>3</sup> 5 7 7 1 <sup>3</sup> 6 7 1 <sup>3</sup> 6 7 1 <sup>3</sup> 6 7 1 <sup>3</sup> 7 7 1 <sup>3</sup> 7 1 <sup>3</sup>	Cm246 4.71 x 10 <sup>3</sup> y a 5 39,5 34 a,9 246 0672	Ст247 25µз 1.6×Ю'у 7,22 а 7,180 7,108 24,707	Cm 248 3.52 x 10 <sup>5</sup> y a 5.08.504 SF g7 248.02	Cm 249 64m <sup>8-9</sup> <sup>7</sup> y~2 E 9	Cm250 1.1 x 10 <sup>4</sup> y SF		Cm252 ≤2d		
Am 240 0.9ms 51h 55 9043, 099, 99, 89,	Am241* 433 y 549,415,551 56 05,17 (mb -90) 56 05,17 (mb -90) 76 010 (mb -90)	Am242 <sup>H1</sup> I6ms 52 y 7 51 11 0 49 67 11 0 49 67 11 0 49 67 11 0 49 67 11 0 49 7 11	Am2435- 7370y 352552344 535 9075 9075	Am24464 9ms) - 11 (0.16	Am245(5/4) 2.04h 8 <sup>90,</sup> 2.036, 252, 240,296, 5.903	Am246 2+ 39m 25.0m 7680, \$1.3,160, 205, 210 154,,7108, 80, 17-174	Am247 22m y0.285,0.226						
Pu239 // 24,390y 4516,515,511 7057,0153 - 77 557,0153 - 77 577,0153 - 777 577,0153 - 777 577,0153 - 777 577,0153 - 777,0153 - 777,0153 - 777,0153 - 777,0153 - 777,0150 - 777,0100 - 777,0100 - 777,0100 - 777,0100 - 777,0100 - 777,0100 - 777,0100 - 777,0100 - 777,0100 - 777,0100 - 777,0100 - 777,0100 - 777,0100 - 777,0100 - 777,000 - 777	Pu 240 6600y 057.512 7.045.04.668 9 7.080.07.08	Pu2415/* 14.3y a 490, 485 y 149, att g, v3840, vidoo	Pu 242 3.87 x 10 <sup>5</sup> y 3.67 x 10 <sup>5</sup> y 3.61 3.61 3.61 3.61 3.61 3.61 3.61 3.61	Pu 24 3 <sup>20</sup> 4.96h 2.56, 49 084.042.012 581 541 541 541 541 541 541 541 54	Pu 244 8 3 x 10 <sup>7</sup> y a 4 59,4 54 5r oy 10 244 115	Pu 245 <sup>(9-4)</sup> B <sup>-</sup> 93,121 y 327,560, 308. 375,028 · 112 σ.~260 ε 1.26	Pu 246 10.85d 8-15, 33 9.044, 027 225 F 40						
Np 238 <sup>2+6</sup> 2 12d 2 <sup>-1</sup> 24, 25, 3 <sup>-1</sup> 44, 26 7 + 2676	Np 23954 2 350 7 645 - 504 6746 - 2766 - 21	1(-)Np240 75m \ 65m 8 <sup>-2</sup> 18, \ 8.89 9043,55,0085 26-162   16 6- 6-	Np 241 16.0m 8-13 9.13.18	<u></u>							156	•	158
6.750 6.750 8123, 15, 90595, 208, 014- 5, 460, 9(x - 35) 5, 414	UI 99.27 SF 4.51 x 10 <sup>3</sup> y 7 048 cy 2.73, cyc., 5mb	U 239 <sup>(5/1)</sup> 235m 8112128 707470435 0311 707470435 0311 707470435 0311	U240 14.1h 8-36 y 044						154				
Pa236 <sup>12m</sup> 8-3.3 E3.4	Pa237 39m 8-230,135, 7.46,92,09- 1.4 E2.30	L 120					152						

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# LIST OF ATOMIC ELEMENTS

• • •		22	3.0		00
Actinium	AC	39	Mercury	мg	80
Aluminum	AI	13	Molybdenum	Mo	42
Americium	Am	95	Neodymium	Nd	60
Antimony	Sb	51	Neon	Ne	10
Argon	Ar	18	Neptunium	Np	93
Arsenic	As	33	Nickel	Ni	28
Astatine	Ał 🖷	85	Niobium	Nb	41
Barium	Ba	56	Nitrogen	N	,
Berkelium	Bk	97	Nobelium	No	102
Beryllium	Be	4	Osmium	Os	76
Bismuth	Bi	83	Oxygen	0	8
Boron	B	5	Palladium	Pd	46
Bromine	Br	35	Phosphorus	P	15
Cadmium	Cd	48	Platinum	Pt	78
Calcium	Ca	20	Plutonium	Ρυ	94
Californium	Cf	98	Pelonium	Po	84
Carbon	С	6	Potassium	K	19
Cerium	Ce	58	Praseodymium	Pr	59
Cesium	Cs	55	Promethium	Pm	61
Chlorine	Cl	17	Protactinium	Pa	91
Chromium	Cr	24	Radium	Ra	88
Cobalt	Co	27	Radon	Rn	86
Copper	Cu	29	Rhenium	Re	75
Curium	Cm	96	Rhodium	Rh	45
Dysprosium	Dy	66	Rubidium	Rb	37
Einsteinium	Ē\$	99	Ruthenium	Ru	44
Erbium	Ēr	68	Samarium	Sm	62
Europium	ξu	63	Scandium	Sc	21
Fermium	Fm	100	Selenium	Se	34
Fluorine	F	9	Silicon	Si	14
Francium	Fr	87	Silver	Ag	47
Gadolinium	Gd	64	Sodium	Na	11
Gallium	Ga	31	Strontium	Sr	38
Germanium	Ge	32	Sulfur	S	16
Gold	Au	79	Tantalum	Τα	73
Hafnium	Hf	72	Technetium	Tc	43
Helium	He	2	Tellurium	Te	52
Holmium	Но	67	Terbium	ТЬ	65
Hydrogen	н	ĩ	Thallium	TI	81
Indium	In	49	Thorium	Th	90
lodine		53	Thulium	Tm	69
Iridium	lr	77	Tin	Sn	50
Iron	Fe	26	Titanium	Ti	22
Krypton	Kr	36	Tungsten	W	74
Lanthanum	La	57	Uranium	U	92
Lawrencium	Lw	103	Vanadium	V	23
Lead	Pb	82	Xenon	Xe	54
Lithium	Li	3	Ytterbium	Yb	70
Lutetium	Lu	71	Yttrium	Y	39
Magnesium	Mg	12	Zinc	Zn	30
Manganese	Mn	25	Zirconium	Zr	40
Mendelevium	Md	101			

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