CURRENT METHODS OF RADIOACTIVE WASTE DISPOSAL

MIRYOUSSEF NOROUZIAN*

*Research Professor, Institute of Engineering, UNAM

RESUMEN

Los fundamentos y conceptos de los fenómenos nucleares han sido revisados en forma simple y resumida a fin de poder ayudar a aquellas personas que estén poco familiarizadas con la física nuclear.

Los efectos de la radiación nuclear sobre la salud se presentan como base y justificación de la preocupación por los desechos radiactivos existentes en el medio ambiente. También se indican las fuentes de desechos radiactivos, seguidas de una discusión de los métodos usuales para manejo y disposición de dichos desechos.

ABSTRACT

The fundamentals of radioactivity are discussed briefly in a simple manner to give the reader a general understanding of the phenomenon. Both acute and chronic health effects from exposure to nuclear radiation are also presented. The main sources of radioactive waste and contaminants are indicated, followed by an in-depth discussion of the merits and pitfalls of the various methods of handling and disposing of radioactive waste currently used.

1. INTRODUCTION

In 1896, Henry Becquerrel was the first person to become interested in radioactivity. He discovered this phenomenon while observing fluorescence and phosphorescence. Later, another scientist by the name of Ernest Rutherford demonstrated the ionization effect of radioactive materials. Madam Curie, working with her husband Pierre Curie, related radioactivity to the atomic structure of the substance.

Radiation has been defined by physicists as the emission and propagation of energy through matter or space by means of electromagnetic disturbances, which display both wave-like and particle-like behavior. Nuclear radiation is that emitted from atomic nuclei in various nuclear reactions and includes alpha, beta, and gamma radiation and neutrons. However, this does not necessarily mean that all three emanations will come from a certain radioactive substance. This simply means that radioactivity involves the transmutation of elements.

An alpha particle is the nucleus of a helium atom, which consists of two protons and two neutrons. It can be easily stopped, for example, by a

sheet of paper. It does not travel more than several centimeters in the air. An alpha particle carries $+2 \times 1.6 \times 10^{-19}$ coulombs electric charge, weighs 4.0026 AMU (rest mass), and travels at velocities ranging from 1.4×10^{9} to 2×10^{9} cm/s (about 10 per cent of the speed of light) in the air.

A beta particle is a high speed electron. It carries -1.6×10^{-19} coulombs electric charge, weighs 5.5×10^{-4} AMU (rest mass) or 9.1×10^{-28} g, and travels at speeds ranging from 30-99 per cent of the speed of light. The penetrating power of the beta particles is about 100 times that of an alpha particle. Therefore, beta particles can travel several hundred feet into the air. Beta particles are emitted with a continuous range of velocities from a nucleus. The basic question concerning beta particles is -Where does it come from when we know there is no electron in the nucleus? According to the law of the conservation of energy and of momentum, emission of a beta particle must be accompanied by the emission of another particle of negligible rest mass and zero charge. This particle is known as a neutrino and the existence of neutrinos has been proven by Reines and Cowan in a series of experiments in 1953 and 1956.

A gamma ray is a high energy form of electromagnetic radiation which travels at the speed of light. This is a highly penetrating ray. For proper shielding from gamma rays several centimeters of lead are needed. There is a question about how gamma radiation involves the transmutation of elements. The answer simply is that gamma radiation *per se* does not cause the trasmutation but that some elements emit alpha particles, others emit beta particles, while gamma rays sometimes accompany one or the other. When for example, alpha particles are emitted from radium they may come in two groups, a group with the energy of 48.79×10^{-5} eV and another group with the energy of 46.96×10^{-5} eV. When an atom of radium emits an alpha particle of low energy, the resulting nucleus will have a higher energy than if a high speed alpha particle has been emitted. If radon (from radium) goes from the excited state to the low energy state, a gamma ray with the energy of $48.79 \times 10^{-5} - 46.95 \times 10^{-5} = 1.84 \times 10^{-5}$ eV must be emitted. Some measure

ment of gamma ray energy from radium shows 1.89×10^{-5} eV which is in good agreement with the above calculated value. When an atom emits an alpha particle, its atomic number and mass number are reduced by two and four, respectively. Emission of a beta particle causes an increase of one in the atomic number. An alpha or beta particle emission from a nucleus is a haphazard event, and hence, in the case of the presence of a large number of nuclei, the emission pattern obeys the law of probability.

In Fig 1 the decay curve for the radioactive element polonium is shown. This curve resembles the time constant curve in electronics (Fig 2). A decay curve is useful for the determination of the half-life of a radioactive atom. If the logarithm of the activity (decay rate) of the atom is plotted as a function of the time, the resulting plot will be a straight line whose slope indicates the half-life of the atom in question.

In the study of radioactivity the following basic questions must be considered and answered (Ref 1):

1. What is the parent nucleus?
2. What particle is emitted from this nucleus?
3. What is the half-life of the parent nucleus?
4. What is the resulting nucleus (products)?
5. Is the product nucleus radioactive, and if so, what are the answers to questions 2, 3 and 4 for this nucleus?

The diagram in Fig 3 gives the necessary answers to these questions.

This diagram shows mass number versus atomic number for uranium. Unit increase of the atomic number with no change in the mass number shows the emission of a beta particle, a decrease of four in the mass number indicates the emission of an alpha particle.

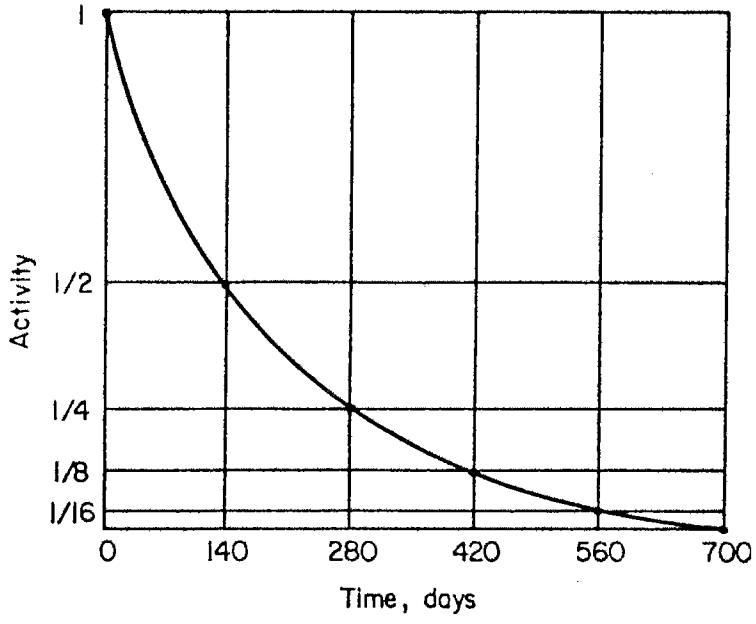


Fig 1. Decay curve for the radioactive element polonium. Polonium has a half-life of 140 (ref 1)

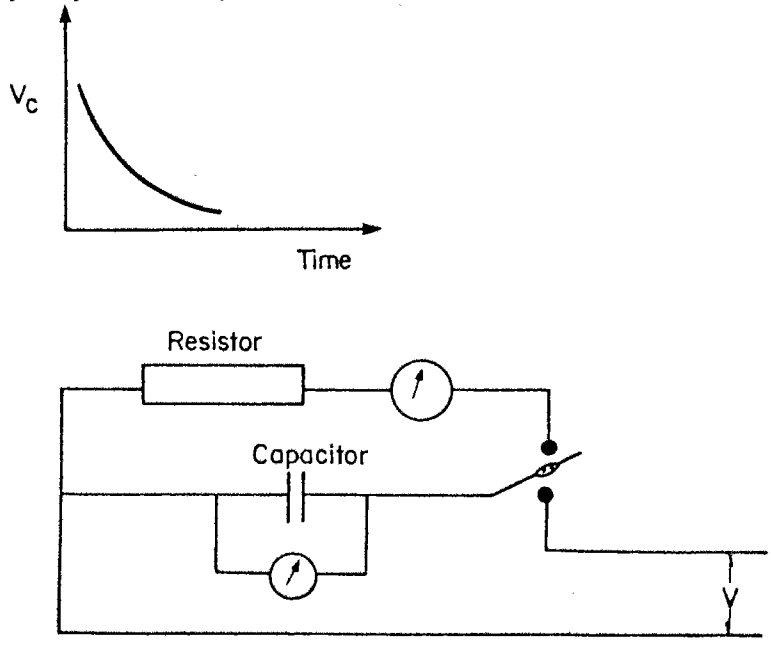


Fig 2. Discharge of capacitor through a resistance

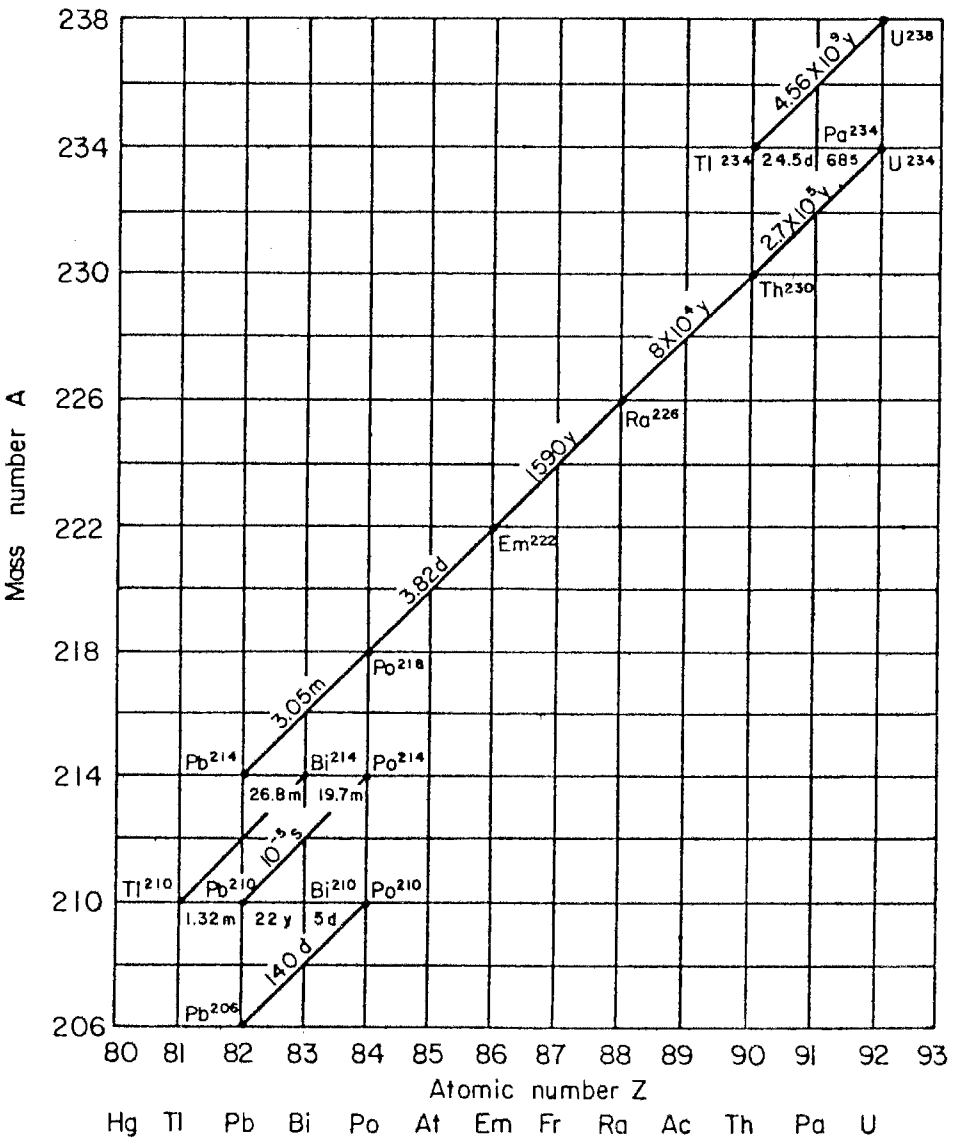


Fig 3. The uranium series of radioactive elements (ref 1)

2. NUCLEAR FISSION

Nuclear fission is the splitting of an atomic nucleus, which is the result of bombardment by neutrons, into two other atomic nuclei each of approximately half the mass of the parent nucleus. Fission is accompanied by the emission of further neutrons and much energy; the process may therefore, under suitable conditions become self-propagating (provided the sample stays together) either slowly, or as in the case of the atomic bomb, in a very rapidly cumulative manner. Many of the fission products are stable, but some are highly radioactive. Four types of fission reactions are very important, these are:

1. Fission reaction by thermal neutrons. Thermal neutrons are those neutrons of very slow speed and consequently of low energy. Their energy is of the same order as the thermal energy of the atoms or molecules of the substance through which they are passing; for example, about 0.025 electron volts which is equivalent to an average velocity of about 2,200 m/s. Thermal neutrons are not effective on ^{238}U, but ^{235}U can be bombarded by thermal neutrons very effectively.

2. Fission reaction by fast neutrons. Fast neutrons are those neutrons resulting from nuclear reaction that have lost little of their energy by collision and therefore travel at high speeds. It is usual to describe neutrons with energies in excess of 0.1 MeV as *fast fission* and in this context the neutrons are so described if they have energies in excess of the fission threshold of $^{238}_{92}U$, i.e., above 1.5 MeV. ^{238}U suffers fission only with fast neutrons.

3. Particle-induced fission. It is possible to induce fission process in elements which have an atomic number above 90 by bombardment with protons, neutrons, and alpha particles. Protons of about 7 MeV are very effective in the bombardment process (Ref 2).

4. Spontaneous fission. ^{238}U undergoes spontaneous fission and in general, the rate of spontaneous fission increases with cross section, indicating a greater probability of penetration of the potential barrier (Ref 2). The half life of ^{238}U for spontaneous fission is about 4.5×10^9 years. Therefore, on the average, 25×10^4 nuclei/g undergo spontaneous fission every hour.

Energy which is released in the fission process has several forms. Distribution of fission energy can be easily approximated by the following table (Ref 1):

Kinetic energy of fission fragments	168 MeV
Energy of fission neutrons	5
Instantaneous gamma-ray energy	5
Beta particles from fission products	7
Gamma-rays from fission products	6
Neutrinos from fission products	10
	201

The gamma-rays, neutrons, and beta particles are absorbed and hence, their energy appears in the form of heat.

The fission fragments distribution has been studied carefully and the frequency distribution of these fragments is shown in Fig 4. In this figure, the mass number of the product is plotted *versus* its fission yield. The reason for choosing mass number instead of atomic number is that these fragments usually decay by ejecting beta particles. Beta particle emission does not affect the mass number. It is obvious from this curve, that the products from fission fall into two mass number groups ranging from 72 to 160.

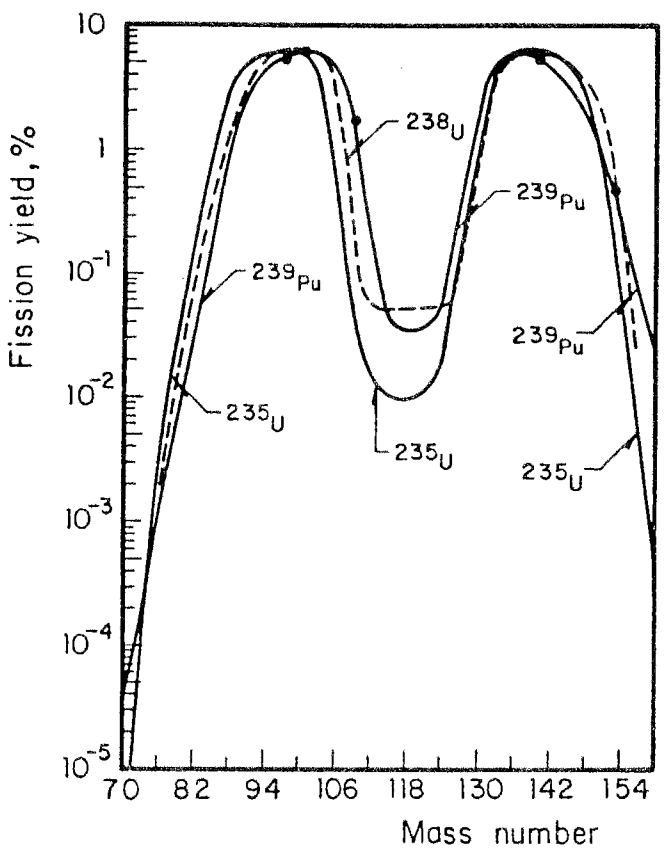


Fig 4. Fission yields in ^{235}U, ^{238}U, and ^{239}Pu; ⊗ fast-fission yields in ^{237}Pu (ref 1)

The fact that the radioactive decay follows exponential law, $R = R_o e^{-\lambda t}$, is strong evidence that this phenomenon is statistical in nature; every nucleus in a sample of radioactive material has a certain probability of decaying, but there is no way of knowing in advance which nuclei will actually decay in a particular time span (Ref 3). If the sample is large enough, that is, if many nuclei are present, the actual fraction of it that decays in a certain time span will be very close to the probability for any nucleus to decay. The statement that a certain radioactive isotope has a half life of five hours then, signifies that every nucleus of this isotope has a 50 per cent chance of decaying in any five hour period. This does not mean a 100 per cent of decaying in ten hours; a nucleus does not have a memory, and its decay probability per unit time is constant until it actually does decay. A half life time of five hours implies a 75 per cent probability of decay in ten hours, which increases to 87.5 per cent in fifteen hours, a 93.75 per cent chance in twenty hours because in every five hours there is a 50 per cent chance (Ref 3).

The empirical activity law, $N_t = N_o e^{-\lambda t}$, follows directly from the assumption of a constant probability of λ per unit time for the decay of each nucleus of a given isotope. Since λ is the probability per unit time, λdt is the probability that any nucleus will undergo decay in a time interval dt. If a sample contains N undecayed nuclei, the number dN that decay in a time dt is the product of the number of nuclei N and the probability λdt that each will decay in the time interval dt. That is, $dN = -N\lambda dt$ where the minus sign is required because N decreases with increasing time t.

3. ARTIFICIAL NUCLEAR DISINTEGRATION

It was in 1919 that Rutherford succeeded in bombarding nitrogen with alpha particles. In doing this, the following reaction took place:

$$^{4}_{2}He + ^{14}_{7}N \rightarrow ^{17}_{8}O + ^{1}_{1}H$$

It shoud be noted that the sum of the initial atomic numbers is the same as the sum of the final atomic numbers, a condition imposed by the conservation of charge law. The initial rest mass is not equal to the final rest mass, but the sum of the initial mass number is equal to the final mass number. Nuclear reaction energy is the difference between the rest masses. Einstein's equation is used for equivalence of mass and energy: $E = mc^2$, where E, m, and c are energy, mass, and speed of light, respectively. If the final sum of masses is less than the initial sum, energy is released in the form of kinetic energy of the final particles.

Example 1.

In the nuclear reaction of $^{4}_{2}He + ^{14}_{7}N \rightarrow ^{17}_{8}O + ^{1}_{1}H$ the rest masses of the particles in AMU (from $E = mc^2$ it follows that 1AMU = 931 MeV) are as follows:

$^{4}_{2}He$ =	4.00260	$^{17}_{8}O$ =	16.99913
$^{14}_{7}N$ =	14.00307	$^{1}_{1}H$ =	1.00783
	18.00567		18.00696

It is obvious that the amount of energy absorbed in the reaction is 1.2 MeV (18.00567 - 18.00696)(931) = - 1.2 MeV.

Example 2.

In the nuclear reaction of $^{1}_{1}H + ^{7}_{3}Li \rightarrow ^{4}_{2}He + ^{4}_{2}He$ the rest masses of the particles in AMU are as follows:

$^{1}_{1}H$ =	1.00783	$^{4}_{2}He$ =	4.00260
$^{7}_{3}Li$ =	7.01601	$^{4}_{2}He$ =	4.00260
	8.02384		8.00520

The sum of the final rest masses is smaller that the sum of the initial values and 17.4 MeV will be released, (8.02384 - 8.00520) (931) = 17.4 MeV.

4. NEUTRONS

In 1930, Becker and Bothe bombarded beryllium with alpha particles. They found that the emitted particles had greater penetrating power than the original alpha particles. Chadwick in England repeated the same experiments and interpreted the results. Based on the characteristics of these particles, he called the particles *neutrons*.

$$^{4}_{2}He + ^{9}_{4}Be \rightarrow ^{12}_{6}C + ^{1}_{0}N$$

The neutrons do not carry any electric charge and hence they do not produce ionization in their passage through gases. They are not deflected by the electric field. The neutrons are stopped only by colliding with the other particles. This collision can be either elastic or penetrating. According to the law of conservation of energy and momentum, if an elastic body collides with a motionless elastic body of the same mass, the first body is stopped and the second body will carry on the motion. Proton and neutron have almost the same mass, and hence, the proton can be used for

slowing down fast neutrons. For this reason, water and paraffin, having an ample amount of hydrogen atoms can be used for slowing down or stopping fast moving neutrons.

5. BREEDER REACTORS

When ^{238}U and ^{232}Th capture neutrons, they do not generally suffer fission; rather, they yield particles that undergo spontaneous changes leading to the formation of fissionable nuclei. In this way, the two inactive species known as fertile materials, are converted into different elements ^{239}Pu and ^{233}U, respectively. These are fissionable and are capable of acting as nuclear reactor fuels. The process is called conversion, and the reactions are as follows:

a) With ^{238}U

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

$$^{239}_{92}U \xrightarrow{23\ min} ^{239}_{93}NP + ^{0}_{-1}e$$

$$^{239}_{93}NP \xrightarrow{23\ days} ^{239}_{94}Pu + ^{0}_{-1}e$$

It is interesting to note, that $^{239}_{94}Pu$, like $^{235}_{92}U$, is fissionable and can be used in nuclear reactors and weapons. Plutonium is chemically different from uranium; its separation from remaining $^{238}_{92}U$ after neutron radiation is more easily accomplished than the separation of $^{235}_{92}U$ from the much more abundant $^{238}_{92}U$ in natural uranium.

b) With $^{232}_{90}Th$

$$^{232}_{90}Th + ^{1}_{0}n \rightarrow ^{233}_{90}Th$$

$$^{233}_{90}Th \xrightarrow{23\ min} ^{233}_{91}Pa + ^{0}_{-1}e$$

$$^{233}_{91}Pa \xrightarrow{27\ days} ^{233}_{92}U + ^{0}_{-1}e$$

The reactor in which fissionable material is produced is called a convertor. If more fissionable material is produced than consumed, the reactor is called a breeder. Breeder reactors are an important and necessary innovation because uranium reserves are limited, making it necessary to convert all of the ^{238}U reserves to ^{239}Pu. Also, the vast reserves of Th can be used by converting fertile ^{232}Th into fissionable ^{233}U.

Breeders are constructed with a blanket of fertile material, ^{238}U or ^{232}Th, surrounding a core of concentrated fissionable material; ^{235}U, ^{239}Pu, or ^{233}U. Extra neutrons rising from the fission of the core material are captured by the blanket of fertile material to form-*breed*-more fissionable atoms, hence, the breeder reactor is a source of energy, and, at the same time, it will produce more fuel than it consumes.

At this point, the superiority of the breeder reactor over fission reactors is obvious. Other advantages of the breeder reactor are outlined below:

The efficiency of breeder reactors could possibly reach as high as 40 per cent

This high efficiency causes less thermal pollution

It uses uranium more efficiently

Uranium and thorium mining is reduced by a factor of at least 1 000

The size and number of processing plants are reduced, this results in much less pollution of all types.

With all of these advantages, breeder reactors do have some drawbacks.

The more important drawbacks inlcude:

1. The misuse of plutonium in bombs
2. Waste treatment and disposal problems
3. Radiation hazards in mining, processing, and reactor sites
4. Breeder reactors are more prone to sabotage.
   Here, it is important to note that 286 kg of enriched uranium have been stolen from the Irvin Atomic Center in the United States since its establishment (Ref 4). This is a very important problem that is common to both breeder and fission reactors.
5. Plutonium is extremely toxic, because it is an α-emitter which can become concentrated in bone. For reasons not clearly understood, it is four to five times more toxic than an equivalent amount of radium, and the permissible body burden is only 0.0005 micrograms.

6. NUCLEAR FUSION

Fusion is the reverse of fission in principle. In fusion, two lighter nucleus are converted into a heavier nuclei. Fusion is the source of nearly all the energy in the universe. This is the main process that occurs on the Sun. Basically, there are two types of fusion-hydrogen cycle and carbon cycle. The proposed fuel for fusion reactors is deuterium which is called heavy hydrogen and denoted by ${}^{2}_{1}H$ or D. The basic reactions are:

$$ {}^{2}_{1}H + {}^{2}_{1}H \rightarrow {}^{3}_{2}He + {}^{1}_{0}n + 3.25 \text{ MeV} $$

$$ {}^{2}_{1}H + {}^{2}_{1}H \rightarrow {}^{3}_{1}H + {}^{1}_{1}H + 4 \text{ MeV} $$

$$ {}^{2}_{1}H + {}^{3}_{1}H \rightarrow {}^{3}_{2}He + {}^{1}_{0}n + 17.6 \text{ MeV} $$

$$ {}^{2}_{1}H + {}^{3}_{2}He \rightarrow {}^{4}_{2}He + {}^{1}_{1}H + 18.3 \text{ MeV} $$

As a result of the first three reactions, five nuclei of deuterium with a total mass of 10 amu will produce an energy of 24.8 MeV. In the fission of ^{235}U, a mass of 235 amu will give 200 MeV of energy. A comparison, based on weight-for-weight, reveals that the fusion process is able to produce about three times more energy than the fission process.

For the fusion process to take place, temperatures of 2×10^6 to 300×10^6 °K are required. At these high temperatures, all the atoms of hydrogen will be ionized and a plasma will be formed. There are at least three ways to heat the plasma; namely, ohmic heating, laser beams, and magnetic compression. A physical container for such a hot gas is out of the question. A magnetic bottle is under investigation for use as a container.

The availability of fuel is the main advantage a fusion reactor would have. Deuterium is present in all water with an approximate ratio of one deuterium per 6,500 light hydrogen in water. In this fashion, one gallon of water would be the equivalent of 300 gallons of gasoline. Separation and processing of deuterium is far easier, safer, and cheaper than fertile and fissionable uranium species. For this reason, the hydrogen cycle is of prime interest to today's researches. Remember that deuterium is not a radioactive element. Radioactive processing sites are the main waste producing sources but this would not be the case with the fusion process. Some other advantages of fusion reactors are summarized below:

1. It is expected that the efficiency of fusion reactors will be in the range of 60-90 per cent vs the efficiency of 33 per cent for today's fission reactors.

2. The thermal pollution that would result from fusion reactors would be far less than that from other types of reactors.

3. Air and water pollution would be negligible when compared to fission reactors.

4. The amount of hazardous waste would be very little.

5. Far less elaborate safety systems are needed in fusion reactors vs a very complicated fail-safe system needed for fission reactors.

6. A fusion reactor could not *run away* as a fission reactor can.

7. Since the threat of theft of fuel for fission reactors either from the processing plant or the reactor site is a very real and dangerous threat, fusion reactors would solve this problem since their fuel source would come from water.

7. RADIATION HAZARDS

The penetration power of the different forms of radiation is a very important factor to be considered. X-rays, neutrons, beta particles, and gamma rays can penetrate quite deeply into the human body. Alpha particles, because of their poor penetration power can be excluded from this category. It is generally believed that the harmful consequences of various radiations are due to their ability to eject orbital electrons from atoms present in various compounds of which the body is constituted (Ref 5). Radiation duration and dose are as important as is the type of radiation. It should be realized that the various cells of the body have different responses to radiation. For example, the tissues which produce blood cells, those which produce sperm or ova, the skin and lens of the eyes, all respond in different degrees to radiation exposure. In general, the faster a tissue grows, the more sensitive it will be to the radiation.

There are two main types of effects of radiation that can be seen in the human body. These effects are known as somatic effects and genetic effects.

1. Somatic effects are effects that occur with the body of the person exposed to radiation. These effects can be either chronic or acute. Fig 5 shows the relationship between radiation dose and the percent of mortality in acute radiation cases. Somatic effects are from two types of exposure, threshold type, or non-threshold type. Usually, the threshold type action is one in which recovery factors operate successfully until sufficient energy is accumulated to prevent the exposed organ from repair. Skin erythemas are an example of threshold type exposure. The non-threshold type action is one in which there is an effect for any dose of exposure. Radiation mutations are an example of this type, for example, mutations on the cellular level. The threshold and non-threshold effects can be shown graphically (Fig 6).

2. Genetic effects are effects which are felt only by the offspring of the individual that has been exposed to radiation. By far, genetic risks are the most important and need to be studied further because of the grave consequences for future generations. Perhaps the best area to study the effects of radiation on human populations is in Japan with the people who were exposed to the bombings at Hiroshima and Nagasaki and their offspring.

All aspects of radiation damage in Japan were studied by the Japanese Atomic Energy Commission. Correlations were found between the distance of a man from the hypocenter of the blast and his sperm count. Of survivors within 1.6 km of the hypocenter, it was found that six months after the bombings 41 per cent lowered sperm counts and digospermia (Ref 7). The Commission also noted that some men remained sterile for years after exposure, but because of the nature of the male teste, fertility returned in 14 per cent of the cases after fifteen years.

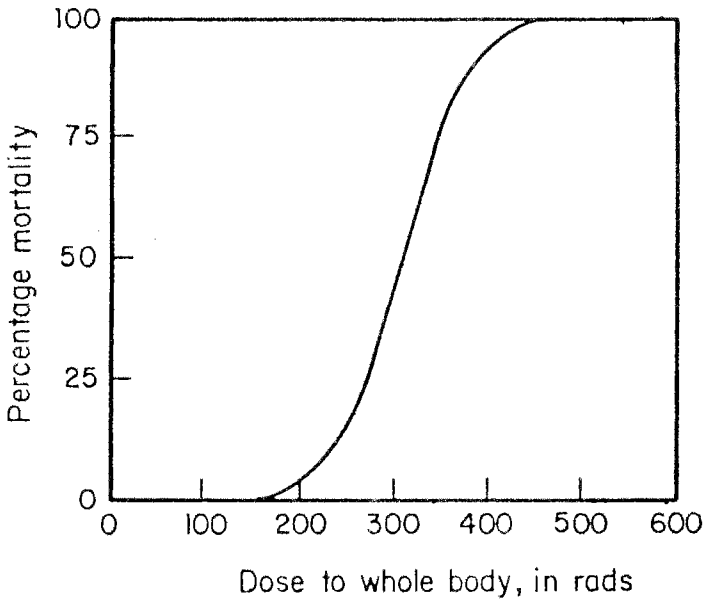


Fig 5. Sigmoid relationship between radiation dose and lethal effect in man (ref 6)

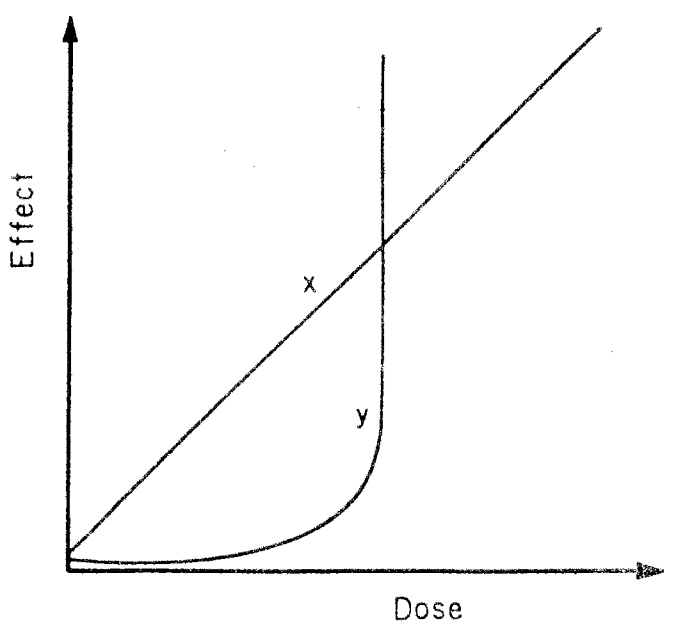


Fig 6. Dose effect curves. Line x is a non-threshold type of response. Line y is a threshold type (ref 6)

Women who were pregnant during exposure to radiation were also studied through termination of the pregnancies. It is not known how many women spontaneously aborted after the bombings, but of those who reached term it was found that in mothers that received more than 20 rads before the child's birth the child was usually born microcephalic; in mother who received over 150 rads before the birth of the child (this was the case in Nagasaki), the child was not only microcephalic but also mentally retarded. The infants were checked every month for nine months after birth and the Commission found the amount of deformities such as blindness, deafness, and mental retardation as well as mutations was not significantly greater than those found in human populations not exposed to the radiation!! (Ref 8).

Now the question is what happens when man is exposed to radiation? When chromosome injury takes place in reproductive cells, they may make phenotypic changes in the first generation. If radiation to the female is extreme, it will result in permanent sterility since all oöcytes are created during the sixth month of fertile life and are never replaced. Males may become sterile but eventually fertility may be restored because of the few living spermatogonial cells of germinal epithelium. However, radiation damage will have occurred.

After the union of sperm and ovum to form a zygote, mitotic division occurs. It is here during anaphase that problems occur. During anaphase, the chromosomes should have paired and split being pulled towards the poles, however, the radiation destroys this process and some chromosomes break and are lost. These never pair correctly again. Telophase follows anaphase in which separation becomes almost complete. The broken chromosomes do not mass near the poles as the normal chromosomes do. This causes the daughter cells to be impaired (Figs 7 and 8).

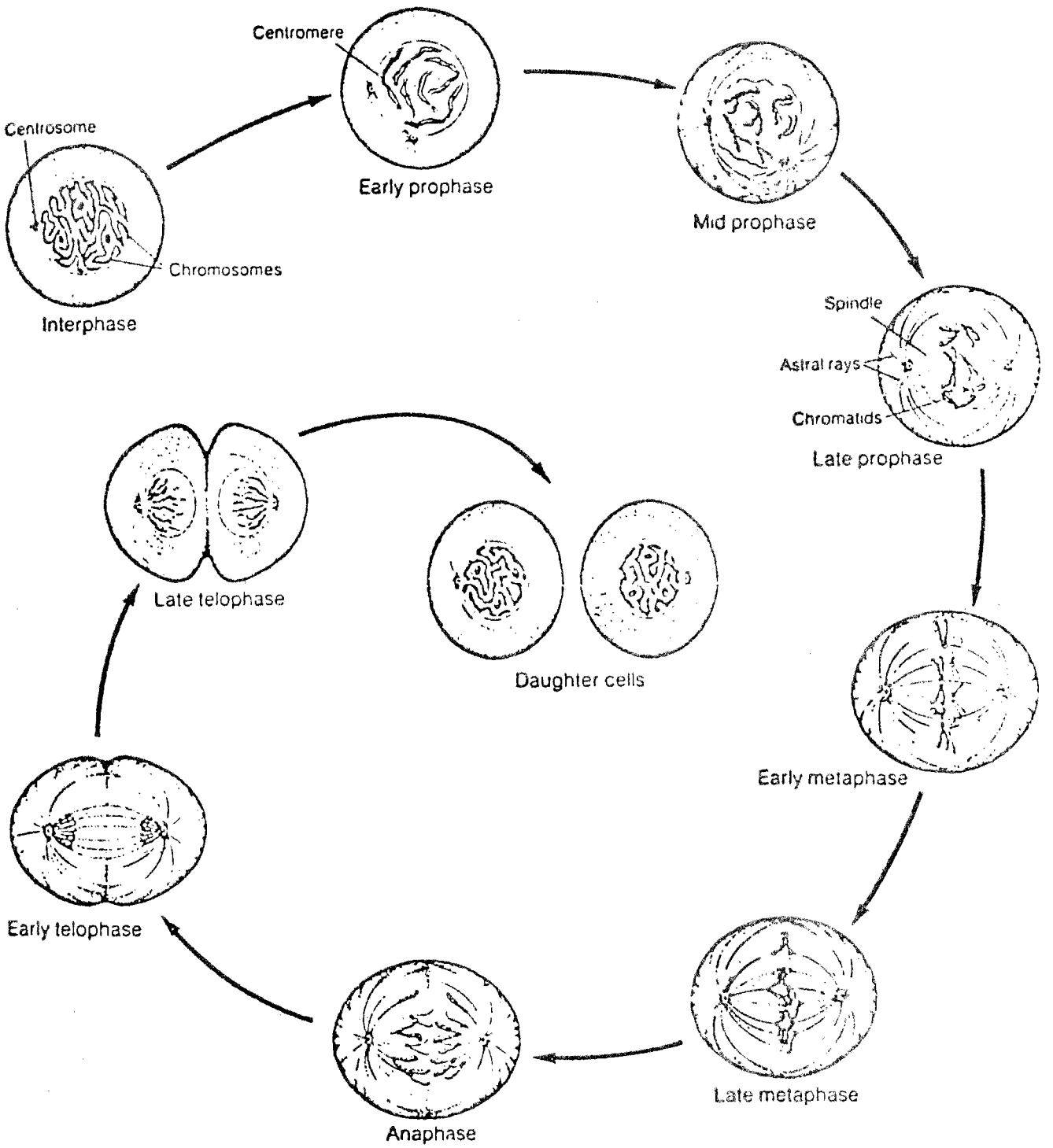


Fig 7. Normal stages of mitosis. The actual living process of mitosis is, of course, a continuous one (ref 7)

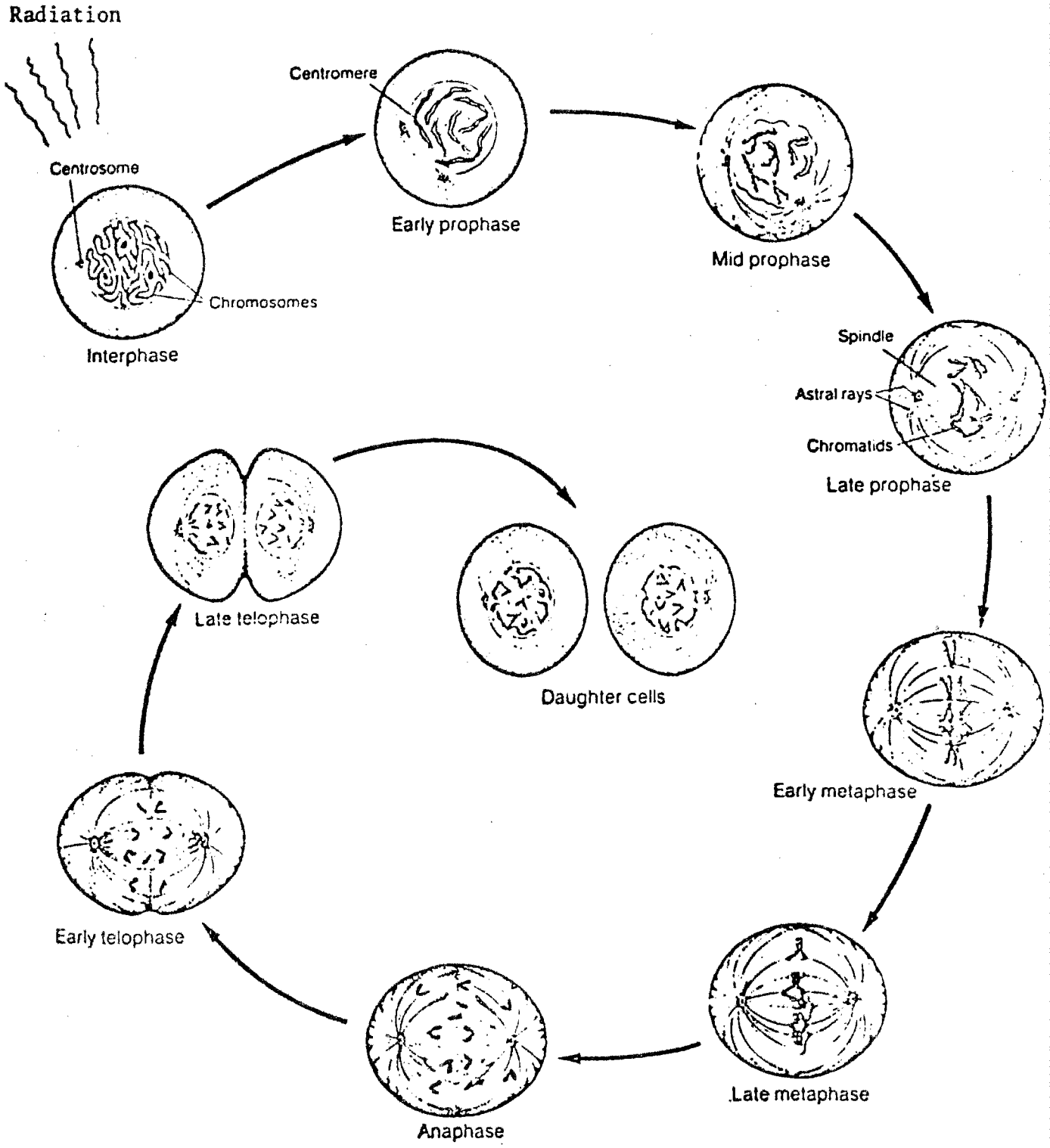


Fig 8. Stages of mitosis after being subjected to radiation (ref 7)

One more point as far as the health hazards of radioactive materials are concerned is the accumulation of radioactive materials in the different parts of living organisms. Some radioactive elements have the same biochemical properties as other elements that are needed by human and animal bodies. Since the body cannot distinguish the active material from the non-active material because of the aforementioned properties, active elements can accumulate in the body and cause very severe health problems. This accumulation can be either direct, for example, by drinking polluted water, or indirect by consuming fish coming from a river receiving radioactive waste products. Table 1 shows some radioactive and non-radioactive elements with similar biochemical characteristics.

Radioactive material inside the body is more dangerous. The reason lies in the fact that α and β particles are not considered hazardous if they are not inside the body, moreover, the body is subjected to continuous radiation when the source of radiation is inside the body.

TABLE 1. SOME RADIOACTIVE ELEMENTS WITH THEIR NON-RADIOACTIVE COUNTERPARTS

Nuclide	Major radiation	Half life	Elements with similar biochemical properties
^{3}H	β - VW	12.3 yrs	H
^{89}Sr	β - S	50.4 days	Ca
^{90}Sr	β - M	28 yrs	Ca
^{129}I	β-W , γ-VW	16×10^{6} yrs	I
^{131}I	β-M , γ-W	8 yrs	I
^{137}Cs	β-M , γ-M	30 days	K
^{140}Ba	β - S	12.8 days	Ca*

S = strong, M = medium, V = very, W = weak, yrs = years,

* not readily absorbed

8. THE SOURCES OF RADIOACTIVE WASTES

A waste is considered radioactive if its radioactive content is more than the maximum allowable level set by the respective authorized organization. Some important sources of radioactive wastes are as follows:

1. Industry: Industries use radioactive material for radiography. Radiography is a method for the inspection of welds and castings. The respective source of radiation is a closed type source. The open type source is used for example to study mixing.

2. Research establishments: This source of radiation includes university laboratories, industrial research centers, and governmental research institutes such as military munitions manufacturing.

3. Hospitals: Hospitals, like industry, use closed and open sources of radioactivity. Closed sources are used for radiography and diagnosis or treatments. Open sources are used for tracing and treatment purposes.

4. Radioactive material processing factories.

5. Nuclear reactors.

Radioactive wastes from all of these sources can be in the form of gas, liquid or solid. Gaseous radioactive wastes cause air pollution, liquid and soluble radioactive wastes create water pollution. Radioactive solid wastes need more careful management. Eventhough its volume is very small when compared to our daily solid waste, its disposal is a much more difficult problem. The following section of this paper will discuss various disposal methods currently under consideration and their advantages and disadvantages.

9. THE DISPOSAL OF RADIOACTIVE WASTES

Before discussing the disposal options available for radioactive wastes, it must be recognized that there are different kinds of radioactive wastes, each having its own disposal problems. Radioactive wastes are divided into three categories: high-level waste nuclides, transuranium isotopes, and very long-lived nuclides.

High-level waste nuclides include such elements as Ru-106 (half-life of 1 year), Eu - 155 (1.7 yrs), Pm-147 (2.7 yrs), Sb-125 (2.7 yrs), Cs-134 (2.1 yrs), Co-60(5.3 yrs), Kr-85 (10.4 yrs), H-3 (12 yrs), Sr-Y-90 (28 yrs), Cs-137 (30 yrs), and Sm-151 (90 yrs). These high-level wastes are legally defined as, *aqueous wastes that result from the operation of the first cycle solvent extraction system, or equivalent and the concentrated wastes from subsequent extraction cycles or the equivalent in a facility for reprocessing irradiated reactor fuels.* These high-level wastes are known for their high heat generation rates and their relatively short half lives (none more than one hundred years).

Transuranic isotopes are defined by the ERDA of the United States as, *any waste material measured or assumed to contain more than the specified concentration of transuranic elements*. The concentration level for transuranic elements is set at ten nanocuries of alpha emitters per gram of waste. Transuranics, also known as actinides, include nuclides that have an atomic number greater than uranium 92. All transuranic or TRU have very long half-lives. Some of those most prevalent in radioactive waste include Np-237 (2.14×10^6 yrs), Np-238 (86 yrs), Np-239 (24,390 yrs), Np-240 (6,580 yrs), Np-242 (3.79×10^5 yrs), Am-241 (458 yrs), Am-243 (7,950 yrs), Cm-245 (9,300 yrs), Cm-246 (5,500 yrs), Pu-238 (86.4 yrs), Pu-239 (24,390 yrs), Pu-240 (6,580 yrs), Pu-241 (13.2 yrs), and Pu-242 (3.79×10^5). Although transuranics are initially less hazardous because of their low concentrations in waste, they are of great concern because of their much longer half-lives than high-level wastes.

The very long-lived waste nuclides, although found in extremely low concentrations, pose serious disposal problems because of the very long half-lives. Some of the very long-lived waste nuclides and their half-lives include I-129 (1.59×10^7 yrs), Tc-99m (2.113×10^5 yrs), Cs-135 (9.5×10^5 yrs), and Se-79 (6.5×10^4 yrs).

Of course radioactive waste comes in liquid, solid and gaseous forms. The majority of today's waste is in liquid form. However, in the future when nuclear power plants are decommissioned there will be much more solid waste in the form of radioactive concrete and other building parts. To date, in the United States alone, there is almost 25,000 metric tons of spent fuel in water storage at commercial nuclear facilities. The U S military has 75,000,000 gallons stored throughout the United States in two hundred underground tanks, and 1,000,000 gallons of transuranic waste has been buried retrievably (Ref 9). It must also be remembered that Japan, Germany, France, England, Israel, Russia, and India also have radioactive waste to dispose of. It is evident from these numbers that a solution to the radioactive waste disposal problem must be found soon.

There are a number of disposal techniques under discussion today. In the past, only two disposal methods were economically and technologically feasible. One method, dilution of the waste in large bodies of water such as the wolrld's oceans is still used by England for disposal of their low-level wastes. This disposal method utilizes the hazard index to justify ocean disposal. The hazard index is defined as *the volume of air or water required to dilute a unit volume of waste or ore to its ingestion radiation concentration guide recommendation* (Ref 10). This disposal method is not a satisfactory alternative to the problem, and the 1977 UN. Law of the Sea Treaty has made ocean disposal of any radioactive waste other than low level waste in a country's national waters illegal (Ref 11).

The second disposal method in use today is not really a disposal method at all, but rather retrievable storage in large tanks until a disposal method is determined. In the United States these tanks are located at the power plant sites and the military reservations that handle nuclear weapons. It must be stressed that this is only a temporary solution to the radioactive waste problem.

The constituents of a typical liquid waste from a nuclear power plant can be found in Table 2. The average activity level for this radioactive liquid ranges from 10-15 curies per liter. The radiant energy of this liquid is so intense, that it causes the liquid to boil. The storage tank must be constructed to stand the corrosive effects of the acid, and the sudden pressure variations. To control the extreme heat in the tanks, they are usually air or water cooled (Ref 12).

Storage has two main problems, one is the lack of storage space after the tank has been filled (many power plants in the United States are running out of tank space already), and secondly, the precarius nature of tank storage. For example, above ground storage tanks are subject to possible earthquake damage, damage during wartime, and of course the most probable

leakage of the tank because of the corrosive effect of the radioactive waste. Tank leakage has occurred in the past and will most likely become more prevalent as the storage tanks age.

In 1973, at the Hanford Reservation in Richland, Washington, over 115,000 gallons of radioactive waste leaked out over a period of 51 days before the leakage was discovered (Ref 13). At this same site, it has been estimated that between August 1958 and June 1973, 422,000 gallons of radioactive waste has leaked (Ref 13).

TABLE 2. TYPICAL LIQUID WASTE CONSTITUENTS

Chemical	Weigh in tons/10,000 m^3
Nitric acid	1,300
Ferric nitrate	7
Aluminum, nickel, chromium and uranium nitrates	5
Mixed fission product nitrates	1.8

9.1 *Solidification*

Solidification is the process of taking liquid waste and changing it into solid form. Solidification of liquid waste reduces the volume of the waste. In the United States it is required that all liquid reprocessing waste must be solidified and disposed of within five years of production. There are three main techniques used for the solidification of waste, these are, calcination, vitrification, and crystalline ceramics.

Calcination is used mainly at the United States military waste reservation in Idaho Falls. In this process, liquid waste is sprayed through an atomizer and dried at very high temperatures. This yields a highly radioactive granular waste known as calcine. The calcine is then placed in drums for storage while waiting further reprocessing.

The most commonly used solidification technique is vitrification. It has been under development for almost thirty years in Europe and the United States. A commercially operating vitrification plant is located in Marcoule, France, receiving wastes for processing from as far away as Japan. Vitrification is a complicated process which takes the product of calcination (calcine) and mixes it with a borosilicate glass frit in a specially made furnace. In the furnace the glass frit and the calcine melt and the waste is then poured into a mold or barrel for storage. A diagrams of the vitrification process can be found in fig 9.

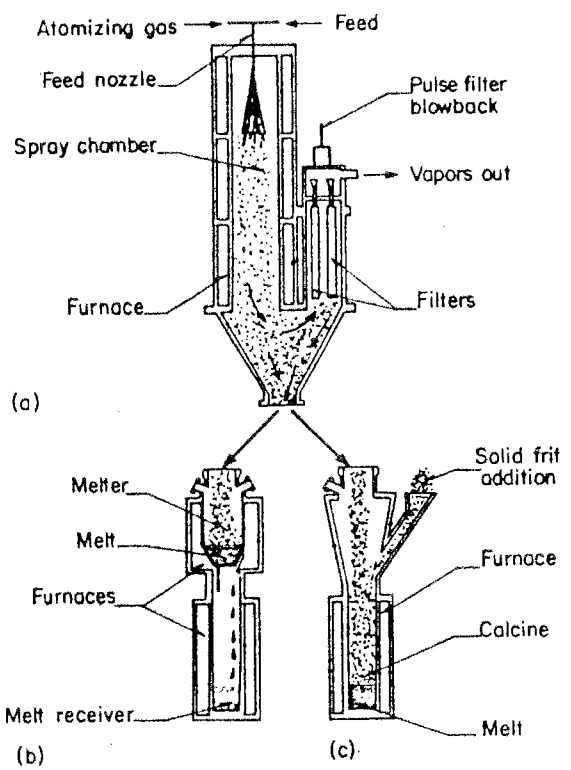


Fig 9. The spray calcination-vitrification process. a) Spray calciner, producing calcine which drops into either, b) continuous silicate glass melter, or c) directly into a waste canister vessel for in-pot melting (ref 14)

Glass is considered by some scientists to be a good matrix for storing radioactive waste since it has strong interatomic bonding and no strict atomic structure. However, in experiments conducted at Pennsylvania University it was found that vitrified waste stored under conditions of severe temperature and pressure (conditions that would be found if the waste was buried) would devitrify over a period of weeks when placed in brine. The waste escaped into solution and formed mineral species not previously present (Ref 15). Some of the new minerals formed however seem to be more stable and insoluble than the original glass. This phenomenon is now under study in the United States and Australia (Ref 12). Because of the instability of the glass waste at extremes of temperature and pressure some scientists feel that a glass matrix may not be the form of choice, but rather ceramics.

Crystalline ceramics are under intensive study for use as a matrix in which to store radioactive waste. Basically, this procedure consists of using a ceramic matrix that will crystallize into an ordered atomic structure suited to specific waste elements. The leachability of this product is low in destilled water, but in brine the leachability is high (Ref 12). The use of crystalline ceramics for waste must undergo much rigorous testing before its full potential is realized.

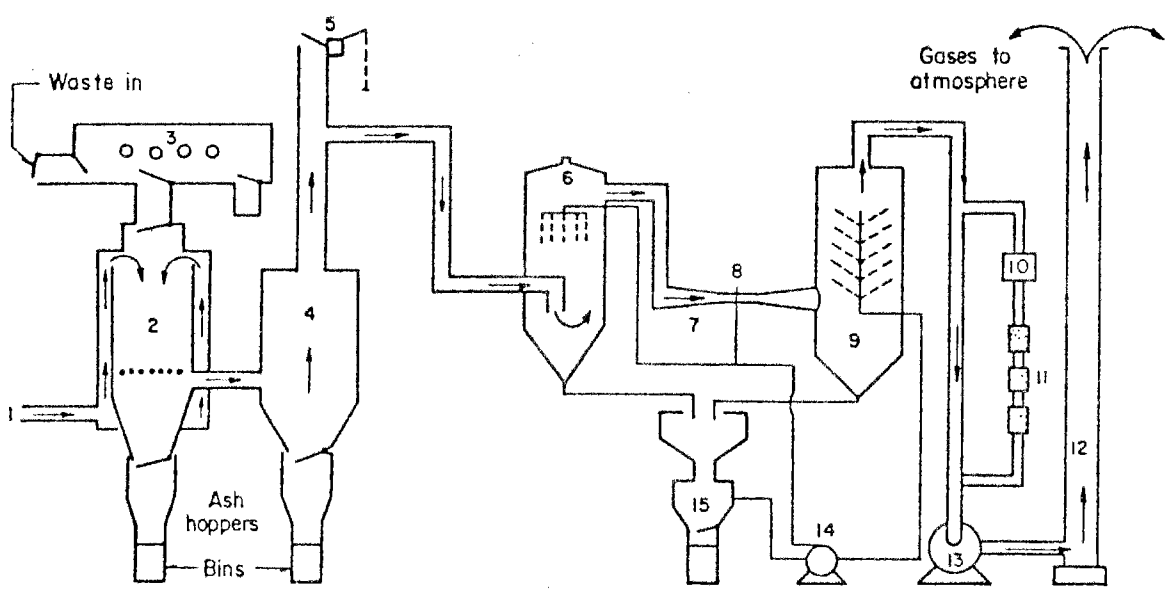
Sometimes incineration is used to dispose of radioactive waste. If incineration is used special attention must be paid to the following requirements:

1. Complete combustion
2. Efficient decontamination of the flue gases
3. Negative pressure within the equipment
4. Ash handling arrangements which do not give rise to a dust hazard
5. Resistent and maintenance free equipment

6. Low conductivity of heat
7. Resistence to chemicals since polyvinyl chloride (PVC) and other chemicals cannot be excluded.

A typical layout for an incineration plant is shown in Fig 10. Dry radioactive ash from an incinerator is not easy to handle. The United States Bureau of Mines has said that the ash hopper must be a disposable container having a solid mass of molten inorganic compound for dissolving active materials. Sodium hydroxide has been suggested as a compound that would meet the necessary requirements.

Experience in the United Kingdom and the United States has shown that incineration of combustible radioactive waste is an economical procedure. A volume reduction by 80 percent can be achieved and the resulting saving in transport, storage and disposal costs is considerable (ref 6).



1. Air intake
2. Incinerator
3. Sorting cabinet
4. Fly ash settler
5. Safety valve flap
6. Adiabatic cooler
7. Venturi scrubber
8. Throat water jets
9. Cyclone scrubber
10. Gas heater
11. Filters
12. Discharge stack
13. Fan
14. Recirculation pump
15. Filter

Fig 10. Typical lay-out for a radioactive waste incinerator (ref 6)

9.2 Geological disposal

The method that is seen as most promising for the disposal of radioactive waste is the placement of these wastes in some type of underground geologic media. The technology exists today for some forms of geologic disposal. This method entails isolating the waste from the biosphere in some geologic formation that is not subject to tectonic activity and the intrusion of groundwater. There have been many suggestions for disposal including placement in mined cavities, matrices of drilled holes, hydro-fracture implacement, deep-well injection, disposal on isolated islands, super-deep holes, rock-melting and mined vaults (Ref 12). American scientists are investigating five types of formations for disposal: basalt, tuff and granite (all of volcanic origin), shale and salt.

9.2.1 Mined vaults. The most discussed technique now is placement of the radioactive wastes in mined vaults. In order to do this, a vault must be dug out of the rock media about two thousand feet below the Earth's surface. The waste materials would be lowered through an access shaft and buried in the floor of the vault. The access shaft would then be back-filled using a material that is impermeable to water and has high ionic retention such as clay.

9.2.2 Super-deep hole. Super-deep hole burial consists of drilling a bore hole at least five miles into the Earth. The waste would then be placed into canisters and stored on top of each other in the hole. It is felt that at this depth there would be no disturbance by surface water, climatic changes or tectonic activity. Because of the extreme pressures at these depths, the hole would seal itself so no media needs to be used to fix the hole.

9.2.3 Rock-melting. A third promising method is rock-melting. In this method the heat produced by the high-level radioactive waste would be used to help store the waste. A deep bore-hole is drilled and liquid waste is pumped into the hole. The liquid, because of the high heat level, would boil eventually boiling dry. During this process it would melt itself into the surrounding rock forming a glassy ball which would permanently immobilize the waste (Ref 16) There are some problems with this technique however, such as the fact that the wastes would be irretrievable, impossible to monitor, and could find their way into the groudwater during the melting period.

Tests done on liquid radioactive wastes stored in trenches and disposal pits at the Oak Ridge National Laboratory found that the ethylenediamine tetraacetic acid (EDTA) which is a commonly used complexing agent in decontamination operations at nuclear plants facilitates low level migration of Cobalt-60 was found in concentrations of up to 10^5 dpm/g in the soil; and 10^3 dpm/ml (450 pCi/ml) in the water. Traces of alpha emitter isotopes of U, Pu, Cm, Th and Ra were also detected (Ref 17).

Hydraulic fracturing, like that used to retrieve more oil from oil wells, is now being used at the Oak Ridge National Laboratory in the United Sates for disposal of radioactive wate. A hole is drilled into a shale formation to a depth of between 1,000-1,500 ft. The hole is then filled with waste and capped with cement. More holes can be cut at slighty higher levels and injections of waste continued (Ref 18). As mentioned above, the waste when disposed of in this manner can migrate into the water and the soil (Ref 17).

All of the rock formations studied to date offer both advantages and disadvantages for use as radioactive waste repositories. One fact must be kept in mind-because a geologic formation has remained stable in the past-does not mean it will do so in the future.

9.2.4 Salt mines. Salt is the geologic media that has been most studied. In fact, Germany is only researching the use of salt mines for disposal (Ref 19). Salt mines do have some helpful properties -salt is plastic and flows under pressure (sealing any openings that are made in the rock), it can dissipate heat, salt occurs in seismically stable areas, and the presence of salt would mean the absence of water. However, salt is not without serious drawbacks. For example, the salt would be highly corrosive to any storage canister, it is highly susceptible to water, heat weakens the structure of salt, it is usually associated with other valuable minerals (salt itself is a valuable mineral), and salt is difficult to monitor.

Basalt is a dense volcanic rock existing in thin layers interspersed with clay materials. Basalt has low permeability and low moisture content, it also remains strong at elevated temperatures. However, Rockwell International was chastised by the Hydrology and Geology Overview Committee (a group of university scientists) for stating that basalt was under study *because of the favorable geology of the site*. The scientists said that the only reason for studying this basalt was the sociopolitical fact that the land is a nuclear reservation (Ref 19).

Granite is a crystalline volcanic rock with low permeability, high chemical stability and the ability to tolerate heat. Granite however is not plastic like salt, that is, it does not flow so it cannot seal cracks and fissures, also, shallow granite formations have many water filled fractures.

Another rock, tuff, formed through volcanic activity is also being investigated. Tuff is formed from the fusion of pumice and ash from volcanoes. Zeolitic tuff has a low density, high water content, and good ionic retention. But, because of the high water content and the fact that tuff is found in geologically active areas subject to serious faulting, the use of tuff as a disposal media is discouraged.

Shale is composed of mud that has become compacted because of pressure. Shale, like salt, is plastic so it flows under pressure, it has low permeability, and high ionic retention. The biggest problem in the use of shale is the fact that it contains large amounts of water which could corrode the waste canisters or contaminate the water table with radioactive waste.

9.3 Ocean Disposal

Ocean disposal of radioactive wastes has come under intense study recently because it has a number of advantages: a) large areas of ocean are available, b) the ocean floor is far remote from any areas of human activity, c) it could provide long term isolation and d) the political difficulties found with geologic disposal would be avoided. However, the constant threat of radioactive waste entering the aquatic food chain eventually to find its way into the human food chain is a threat that must be studied in detail. It would also be next to impossible to retrieve the waste should a problem arise.

Laws also govern the uses of the world's seas and as article 25 (Ref 1) of the First UN Conference on the Law of the Sea (1958) states, *every state shall take measures to prevent pollution of the seas from the dumping of radioactive waste, taking into account any standards and regulations which may be formulated by competent international organizations* (Ref 11). The Third UN Conference on the Law of the Sea (1973-1977) declared that ocean dumping of radioactive wastes in international waters was illegal. However, it was not determined if sub-seabed disposal should be classed as dumping. It was decided that the responsibility should fall to the IAEA. This responsibility includes:

1. Establishment of safety standards and publication of recommendations on radiological protection and waste management.

   a) Legal and administrative measures for control of radioactive waste disposal into the seas and oceans

   b) Studies of the effects of radioactivity on aquatic organisms and ecosystems, as well as the interaction of radioactive contaminants with constituents of the marine environment

   c) Development of guidelines for geologic disposal of high-level and alpha-bearing wastes

2. Recommendations of the safe transport of materials

3. Regional training and technical assistance

   a) Research contracts and agreements on marine science and pollution

   b) Special missions on siting of nuclear facilities on seacoasts and offshore.

Three methods for sub-seabed disposal have been suggested by scientists thus far. These inlcude the burial of wastes in deep ocean trenches, deep ocean sediment burial, and burial in subsediment bedrock.

Deep ocean trenches are found in all of the world's oceans where two tectonic plates meet. It has been suggested that waste canisters dropped into these trenches would, through tectonic activity of the plates, be pulled towards the Earth's center. However, the movement of the waste canisters would be quite slow, leaving the canisters exposed during their most dangerous period.

In deep ocean sediments, found between 2-3 miles below the ocean's surface the waste canisters could be buried in the sediments through the use of a free-fall ballistic penetrometer or the waste could be lowered by cables

into bore holes. The use of cables would allow for retrieval of the wastes should that be necessary. All of the methods must be studied in great detail before any decisions concerning the dumping of radioactive waste in the ocean can be made.

9.4 *Polar disposal*

Disposal of radioactive waste in the Polar ice caps was first suggested by Bernard Philbirth who patented his idea in 1958. There are some positive features to this disposal option. First, the Polar ice caps are well isolated from human contact, ice would provide a good media in which to dissipate heat, the wastes could melt themselves into the ice making drilling unnecessary, and ice is impermeable to water.

There have been various methods proposed for radioactive storage in ice. One method, known as surface storage, would require the wastes be placed in a building constructed on the surface for this purpose. Over time, the building would be covered with snow and ice.

The most popular suggestion is the melt down of the wastes into the ice. This can take two forms; either the waste can be allowed to melt down freely, or it can be held at a certain depth by anchors at the surface of the ice. Melt down does have some drawbacks though. The Polar ice caps are not geologically permanent features and it cannot be guaranteed that the ice caps would remain frozen for the extremely long periods of time necessary for isolation. The temperature limits in the ice are very narrow and the heat from the waste could cause melting. Fresh water lakes are also located under the ice caps and it is possible that the waste could melt itself into one of the lakes. It is also difficult for people to work in the extreme Polar environment. The overriding problem however, is that by international treaty the Polar ice caps cannot be used for the disposal of nuclear waste (Ref 16).

9.5 *Space disposal*

In the United States it has been proposed that radioactive wastes could be rocketed into the Sun or into orbit into space. This system would require the US Space Shuttle to take the waste into space, and from space to fire the waste into orbit.

There are numerous drawbacks to the proposal not the least of which is the cost. It has been estimated that this disposal method would add at least 50 per cent to the average electric bill in the United States (Ref 12). It would require a fail-safe packaging that could survive re-entry and landing on the Earth's surface, or an accident during the launching of the Space Shuttle. Despite these problems, NASA and the Pentagon are seriously considering this method.

10. CONCLUSIONS

Radioactive materials cannot be destroyed by applying any of the chemical or physical procedures available today. Radioactive atoms decay in a fixed and constant rate. The rate of decay varies from element to element. Radioactive materials with longer half-lives are more dangerous because of their long half-lives and hence, careful and long term observance is required. There is only one method known thus far for dealing with radioactive wastes and that is storage (delay-decay). Radioactive waste storage may pass from generation to generation with all of its related history and documentation until it becomes possible to discharge the waste making use of the policy *dilution is the solution to pollution* however, this outcome is highly unlikely given the extreme lengths of time necessary for storage.

Radioactive material is a good servant but a bad master. All efforts must be made to keep it as a servant. There is considerable work concerning the use of radioactive materials but much less research has been conducted concerning the handling of the resulting radioactive wastes. The use of radioactive material and the disposal of radioactive wastes must be equalized in order to prevent having a bad master, radioactive waste around.

Like it or not, as the time passes more radioactive material enters the energy field and gives the signal, *radioactive waste disposal problem* to man.

11. REFERENCES

1. Sears, F W and Zemandky, M W, *University Physics*, 3rd edition, Addison-Wesley Publishing Co, Inc, Reading, Mass (1965)

2. Lapp, R E and Andrews, H L, *Nuclear Radiation Physics*, 2nd edition, Prentice-Hall, Inc, Englewood Cliffs, New Jersey (1958)

3. Beiser, A, *Concepts of Modern Physics*, 2nd edition, McGraw-Hill Book Co, New York (1973)

4. *Islamic Republic (daily newspaper)*, "Robbery of nine kilograms of enriched uranium", Teheran, Iran (monday, june 14, 1982), (in Persian)

5. Glasstone, S, *Sourcebook on Atomic Energy*, 2nd edition, D. Van Nostrand, Inc, New York (1958)

6. Collins, J C and Sir Alexander Fleck, *Radioactive Wastes: Their Treatment and Disposal*, John Wiley and Sons, New York (1960)

7. Winchester, A M, *Genetics: A Survey of the Principles of Heredity*, Houghton Mifflin Co, New York (1972)

8. "Journal of Radiation Research", *Japanese Atomic Energy Commission*, Vol 16-17, Tokyo, Japan (1975/1976)

9. Carter, L, "Radioactive wastes: Some urgent unfinished business," *Science*, 195(4279) (1977), 661

10. Eicholz, G G, *Environmental Aspects of Nuclear Power*, Ann Arbor Science, Ann Arbor, Mich (1976)

11. Deese, D A, *Nuclear Power and Radioactive Waste*, Lexington Books, Lexington, Mass (1978)

12. Lipschutz, R D, *Radioactive Waste: Politics, Technology, and Risk*, Ballinger Publishing Co, Cambridge, Mass (1980)

13. Gillette, R, "Radiation spill at Hanford: The anatomy of an accident", *Science*, 181(4101) (1973), 728-730

14. *McGraw-Hill Encyclopedia of Environmental Science*, Parker, S P, editor, 2nd edition, McGraw-Hill Book Co, New York (1980)

15. McCarthy, G J, *et al*, "Interactions between nuclear waste and surrounding rock", *Nature*, 273 (1978), 216

16. Kubo, A S and Rose, D J, "Disposal of nuclear wastes", *Science*, 182 (4118) (1973), 1205-1211

17. Means, L J *et al*, "Migration of radioactive wastes: Radionuclide mobilization by complexing agents", *Science*, 205(4347) (1978), 1477

18. De Laguna, W, "Radioactive waste disposal by hydraulic fracturing", *Nuclear Safety*, 11 (1970), 391

19. Carter, L J, "The radwaste paradox", *Science*, 219 (1983), 33-36