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SCIENCE

High-Level and Long-Lived Radioactive Waste Disposal

Ernest E. Angino

Summary. No uniform international approach for handling the problem of high-level radioactive waste disposal exists. All the while, the volume of these wastes continues to grow. The only viable solution to the disposal problem is a geologic one. Burial of these wastes in solid form for long periods of time in mined cavities in salt or Precambrian crystalline rock formations is technically possible. Several steps in the burial process have already been demonstrated in Germany. The problem becomes more serious as the number of countries committed to the use of nuclear energy grows. If one considers the problems of seismic stability and worldwide distribution of salt deposits, the overwhelming need for an international solution to the waste disposal problem seems obvious.

No uniform international policy of nuclear waste management exists. Policies completely acceptable in one country are not so in another; what is high-level waste in one country is not so in another. Nevertheless, as the demand for energy increases and the use of nuclear energy expands, the amount of nonrecyclable wastes that are generated will inevitably increase in various parts of the world. Figure 1 and Table 1 portray the trends. As of 1 June 1976, there were 41 countries committed to the use of nuclear power to generate electricity. Of these, 20 countries already produce electricity by nuclear power. Presently, Sweden, one of the early users of nuclear energy, is in the process of reducing its dependence on nuclear generated electrical power. Recent newcomers to the list of countries committed to nuclear energy are Turkey and Indonesia. In addition, five other countries have reported long-range plans to build nuclear power plants: Cuba, Kuwait, Libya, New Caledonia, and Peru. A detailed list of countries with reactors operating, under construction, or planned is given by the International Atomic Energy Agency (*1*).

At present, the unusable fraction of high-level radioactive wastes that has accumulated from operating reactors and military uses is being stored above ground in liquid form in various countries of the world. If the long-term security and safety of man is desired, these wastes must be removed from the surface of the earth. The logical solution, then, is burial of the wastes in a geologic formation of great integrity. The only viable alternative to this would be 100 percent recycling of *all* the wastes, but this does not appear to be technologically feasible at present. Unfortunately, this problem is not new. We have been warned of its seriousness in the past but have chosen to ignore it until recently. Sterling Cole (*2, 3*) commented at least 15 years ago that "The method of reactor waste disposal in any country concerns the welfare of all nations." And Ralph Lapp (*4*) stated: "It is important that nuclear power risks be placed in perspective since a society dependent on high technology necessarily abandons the concept of zero risk."

High-Level Radioactive Wastes

High-level wastes are defined (*5*) as "aqueous raffinates that result from the operation of the first cycle solvent extraction system, or equivalent, and the concentrated wastes from subsequent extraction cycles, in a facility for reprocessing irradiated reactor fuels." These wastes contain essentially all the nonvolatile fission products, 0.1 to 0.9 percent by weight of the uranium and plutonium originally in the spent fuels, and all the other actinides formed by transmutation of the uranium and plutonium in the reactors. Plutonium and uranium are segregated to recover their fissile values for reuse in the nuclear fuel cycles. Present federal regulations require that these wastes be solidified within 5 years after generation. The resulting stable solids are to be shipped to a federal repository within 10 years after separation of the fission products from the spent fuel. Plans also call for interim storage and isolation of the wastes from man and his environment until ultimate (final) geologic disposal is accomplished.

High-level radioactive wastes can be divided into two categories. One, those fission products of intermediate atomic weight (for example, strontium-90 and cesium-137) and relatively short half-lives ($t_{1/2}$), that is, 30 years or less. This implies that in 700 years, less than 0.0000001 percent of the wastes will remain and they will no longer be a problem. Two, those wastes referred to as the actinides, such as plutonium, neptunium, and americium. Typically, these have long half-lives, for example, the $t_{1/2}$ of ^{239}Pu is 24,400 years. Neptunium has a much longer half-life. These wastes are very toxic (radiogenically and carcinogenically) and will last or be cause for concern for 500,000 to 1,000,000 years if just stored (*6*).

Present waste extraction processes provide for 99.5 percent removal of plutonium and uranium. If we were to require 99.9 percent removal for plutonium, uranium, and neptunium, and 99 percent for americium and curium, the

The author is professor of geology and civil engineering at the University of Kansas, Lawrence 66045.

Table 1. Nuclear commitments in countries other than the United States (25).

Nuclear generator	1974 Capacity (Mw)	1974 Reactors (No.)	1975 Capacity (Mw)	1975 Reactors (No.)	1976 Capacity (Mw)	1976 Reactors (No.)
Operable	24,293	96	29,175	102	35,882	112
Under construction	50,097	77	59,767	85	85,182	117
On order	56,112	73	54,462	70	53,787	58
Planned	90,073	102	150,874	169	168,504	167
Total	220,575	348	294,276	426	343,355	454

long-term activity would be reduced by a factor of 100. The extracted actinide products could then be recycled and burned in the reactor at only a modest loss in efficiency. The waste problem would then become of concern for only 700 to 1000 years—a time period much more tractable from man's frame of reference. This is also a period for which it would be much easier to assure the complete safety and isolation of the wastes than for a million years (5).

The geologic disposal of high-level radioactive wastes raises several questions that must be answered: (i) Can high-level radioactive wastes be stored (isolated) safely underground for long periods of time? (ii) What geologic medium is the safest for long-term burial? (iii) Can the waste be properly stored so that it does not contaminate man's environment? (iv) In what form should the wastes be stored—liquid or solid? (v) Can the wastes be stored in such a manner that they will be retrievable at some future date, if the need arises?

Several steps in the burial process for medium- and high-level radioactive wastes have already been demonstrated in Germany and the United Stated where burial of such wastes is under way or is in the planning stage. Extensive planning and research related to the handling and burial of high-level wastes is in progress in France, Canada, Great Britain, Russia, the Netherlands, West Germany, and other countries (7, 8).

Geologic Disposal

A summary of geologic waste disposal methods and the technological feasibility of each is given in Table 2. The three methods—storage in geologic formations, in ice sheets, and in the seabed—all require considerable amounts of research to assess properly the permanence of the burial, the safety and environmental effects, the retrievability, and long-term (that is, tectonic) stability of the burial areas.

Detailed assessments of each method have been made by Schneider and Platt (9) and the Energy Research and Development Agency (7). Storage of wastes in solid form is most likely. In liquid form the wastes are more mobile and hence could easily move into and contaminate the environment. They are also more difficult to store in liquid form. Systems for converting all the high-level radioactive liquid wastes to an insoluable glass-like solid, a ceramic-metallic form, or bitumenized solid have been proposed and are being tested; the glass-like solid is the safest form at present. This solid would then be placed (permanently or retrievably) in some type of subsurface cavity (mine, vault, or drill hole, for example) in an appropriately deep, stable geologic formation of carefully chosen composition, such as salt, dolomite, or anhydrite.

All these methods have several aspects in common. (i) Interim liquid waste storage at a reprocessing plant is not needed. (ii) Partitioning of the wastes into two or more fractions is not needed. (iii) Wastes for each burial method can be solidified into a boro-silicate glass or other solidified form after placing the wastes in a stainless steel container. (iv) Interim storage of the solid waste is optional. (v) Each requires transportation of wastes to a disposal site or a recycling facility. (vi) Each involves permanent disposal of waste constituents. (vii) Each involves total disposal of high-level waste constituents.

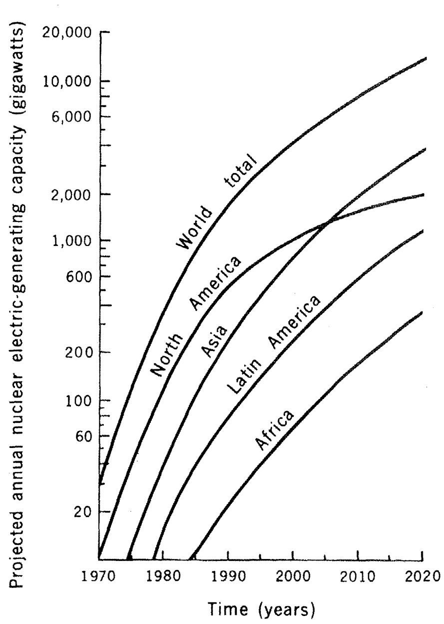


Fig. 1. Projected worldwide nuclear power generating capacity by continent. [Modified from Epstein (3)]

Of the methods listed in Table 2, those that have the most to recommend them, considering geologic, safety, and isolation criteria, are solid waste emplacement in a mined cavity (Fig. 1) or in man-made structures in a mined cavity. If geographical as well as geological isolation are to be the main criteria, emplacement in mined cavities in the ice-free areas of Antarctica should be considered. Other solid waste burial schemes are illustrated in Figs. 2 to 5.

Some of the factors that need to be considered in selecting a rock formation as a burial site are as follows:

1) The rock formation must have a relatively wide distribution, availability, and horizontal extent, so that it is unlikely to be used as a mineral resource in the future.

2) The rock unit must have a high structural strength in terms of compressibility; good thermal conductivity; and high heat capacity.

3) Knowledge of the groundwater hydrology of the rock formation (velocity, direction of flow, and volume) must be obtained.

4) The site should be in a zone of low seismicity and corresponding high tectonic stability.

5) The containing rock unit should be relatively impermeable; its porosity must be known and evaluated.

6) The rock unit should be relatively undisturbed structurally.

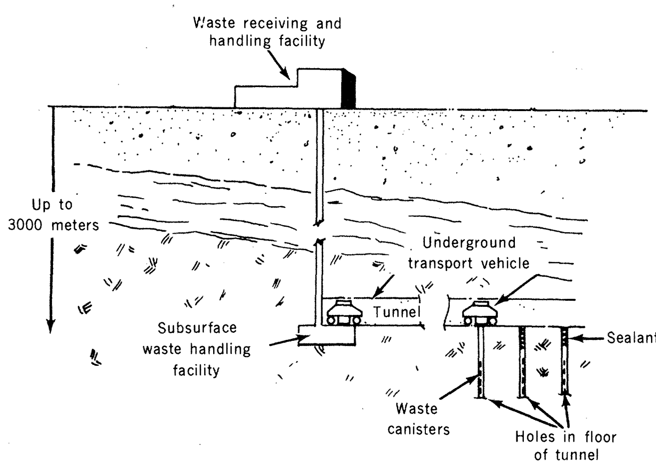
7) The unit should be thick enough for wastes to be buried a minimum of 1000 feet (300 meters) below the surface—and preferably deeper.

8) A monomineralic or homogeneous rock unit should be preferred.

9) The rock unit should have a reasonable plasticity to allow healing in case the burial site is breached.

The most important factor in the containment capability of any burial area is the absence of subsurface water in any form. Water coming into contact with the waste would accelerate the migration of radionuclides. To determine the safety of a particular site we would have to be able to predict the degree to which water might contact the waste following burial, the pathways of water away from the burial site, the ion exchange or absorptive capability of the geologic materials containing the waste and of the rock types along any pathway, and the extent to which off-site subsurface waters might be affected by the burial site. Detailed studies would have to be made in the burial area of the surface and subsurface geology, hydrology, geophysics (thermal effects), and radiation effects on the country rock used for storage. These data would have to provide information on nuclide movement in the area under consideration under any anticipated chemical and physical conditions likely to prevail over a few thousand years.

The rock volumes needed to contain high-level radioactive wastes are not large. For comparison, the total high-level nuclear power plant waste expected to be produced by the year 2000 (in the United States) is about 29×10^6 gallons ($\sim 110 \times 10^6$ liters). One metric ton of processed uranium yields about 1100 gallons of high-level wastes; solidification reduces the liquid volume by about eight times. This amount of solidified waste would cover an area about the size of a football field to a height of 12 to 13 feet (4 m) (*10*). Additional rock volume would be needed to bury the wastes generated by the U.S. military. In 1973 approximately 85×10^6 gallons ($\sim 325 \times 10^6$ liters) were stored by the military and were awaiting solidification (*11*). Burial of these existing wastes would require at least three times the rock volume of the commercial high-level radioactive wastes predicted to be generated over the next 25 years.



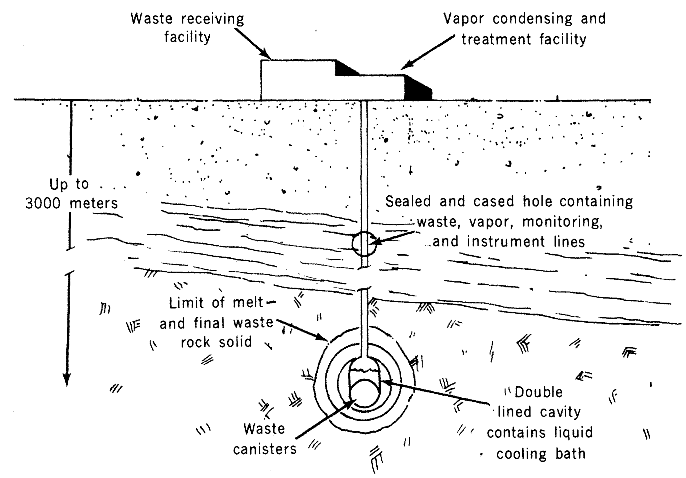


Fig. 2 (left). Solid waste emplacement in a mined cavity with no fluid cooling or melting. [From Schneider and Platt (*9*)] Fig. 3 (right). Solid waste emplacement in a mined cavity with interim liquid cooling and waste rock melting. [From Schneider and Platt (*8*)]

Table 2. Summary of the technical feasibility of alternative nuclear waste management systems. Favorable characteristics include: fair distance from man's environment; safety from storms and most of man's activities. Unfavorable characteristics include: some potential for penetration by man in future; poor retrievability and monitoring; possible groundwater transport. Differences from these general points are indicated to right of each method under the appropriate column. [Modified from Schneider and Platt (*9*)]

Storage method	General characteristics relative to feasibility	
	Favorable	Unfavorable
	Storage in geologic formations	
Solid waste emplaced in mined cavity; no fluid cooling or melting	Ion exchange of rocks as backup	
Solid waste emplaced in mined cavity; initial water cooling; melting*	Ion exchange of rocks as backup	Irreversible high temperature in rock
Solid waste emplaced in man-made structure in mined cavity; initial air cooling; no melting	Ion exchange of rocks as backup; provides ready interim retrievability	Requires interim operation by man
Solid waste emplaced in man-made structure in mined cavity; initial water cooling; no melting	Ion exchange of rocks as backup; provides ready interim retrievability	Requires interim operation by man
Solid waste emplaced in matrix of drill holes; no fluid cooling or melting	Ion exchange of rocks as backup	Very poor retrievability and monitorability; many penetrations to surface
Solid waste emplaced in deep holes; no fluid cooling; melting or nonmelting†	Ion exchange of rocks as backup; large distance from man's environment	Very poor retrievability and monitorability; deep geology unknowns
	Storage in ice sheets	
Self melt through ice‡	Great distance from man	Extended transport; poor retrievability
Anchored storage or disposal‡	Low temperature for cooling	Extended transport
Ice surface storage or disposal‡	Possible international solution	Many technical unknowns
Antarctica (subsurface burial in ice-free areas)	Great distance from man; low temperature for cooling; possible international solution	Retrievability and monitorability good
	Storage in the seabed	
Subduction zones and other deep sea trenches†‡	Great distance from man; water for dilution	Extended sea transport; mobility of seawater
Stable deep sea areas	Ion exchange of sediments as backup	Concentration by ecology
Rapid sedimentation areas‡	Possible international solution	Very poor retrievability and monitorability

*This method can also involve in-place melting and conversion to a rock-waste matrix. †Cannot be implemented with today's technology. ‡These have an uncertain potential for providing adequate safety.

Thermal Characteristics

Regardless of the geologic material (for example, a monomineralic unit such as salt or anhydrite) or rock type in which burial is to take place, it is essential to have detailed knowledge of the heat capacity and thermal response of these materials to the thermal energy of the waste to be contained therein, because many rock properties are temperature-dependent. The rock materials in which the waste container is buried should dissipate heat rapidly, yet remain stable themselves while at elevated temperatures. Many rock properties are also radiation-dependent. That is, they are altered depending on the amount of radiation to which they are exposed. Detailed studies of the radiation and thermal variations expected with time upon exposure to radioactive wastes have been made for salt (*12*) but appear to be lacking for most other rock types that might serve as burial chambers for the waste.

Data of this type are essential because transport of radionuclides in geologic media is a complex function of (i) the physical, chemical, and radiological properties of the medium, (ii) the physical and chemical form of buried radioactive materials, and (iii) the driving force for nuclide migration, which can be water flow, temperature differences, or concentration differences.

Recent studies (*13*) have shown that the migration rate of americium and plutonium in relation to water is slow. Migration could be delayed or prevented by appropriate geologic materials so that the intrusion of actinides into the surrounding environment from a properly designed site would be unlikely.

The nuclide migration problem is critical. Some assurance for the limited migration of ^{239}Pu and other actinide species in a specific geologic environment comes from studies of the Oklo natural reactor (*14*). There the data show that the escape of radioactive products from the reactor zone after almost 2 billion years has been very limited. While it is not possible to draw precise conclusions about the storage of radioactive wastes from studies of the Oklo phenomenon, the data are reassuring.

A major problem related to the migration of fluid inclusions in different rock or mineral materials is the tendency for the fluid to move up the thermal gradient—eventually reaching the heat source. This phenomenon acts to bring water into contact with the radioactive waste material. This problem, of course, is more critical for salt than for other rock materials.

The question of thermal conductivity, along with that of the storage of radiation energy in rock adjacent to the buried wastes, deserves far more attention than it has received. Radiolysis and hydrolysis effects of nuclear wastes also require study. Only salt has been adequately studied as regards these effects. We know next to nothing of the detailed thermal, radiation, radiolysis, and hydrolysis effects of these wastes on granites, basalts, clay minerals, limestones, dolomites, or anhydrites.

Seismic and Structural Stability

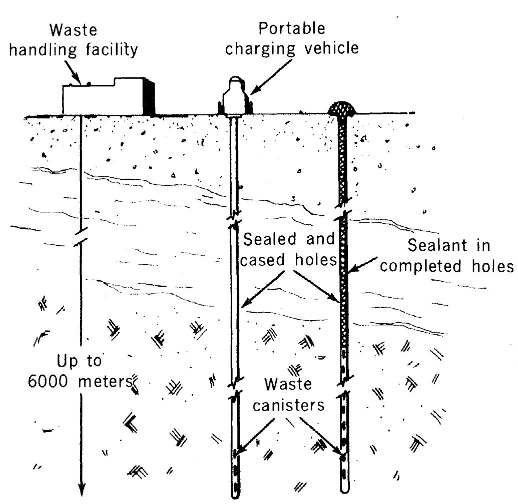
Because high-level radioactive wastes will require burial in areas of low seismic activity and high structural stability, countries such as Japan, Indonesia, and Peru, and islands such as New Caledonia will have to export their wastes to other countries or seek other means of disposal. Those countries with a stable geologic environment may not be willing to accept for burial large amounts of nuclear waste generated in other countries.

The idea of structural stability implies the unlikelihood of the breaking of any burial chamber by way of jointing or faulting, and thus rules out many parts of the United States from consideration as burial sites. A study conducted by the National Academy of Sciences (*15*) concluded that salt (NaCl) is the most desirable rock type in which to bury high-level radioactive wastes for extended time periods. Salt is approximately equal to concrete in its ability to shield harmful radiation. A possible substitute with similar properties might be the potash deposits (sylvite-KCl) of the world. But whereas the use of salt deposits for burial would be unlikely to create a shortage of this mineral resource in the future, potash deposits are in shorter supply and are important as sources for chemical raw materials and fertilizers.

Another alternative being investigated is the use of massive clay deposits (*16*). The thickness, plasticity, large lateral extent, low permeability, high ion exchange capacity, and other properties of clay would ensure preservation of any escaping waste in event of subsequent faulting. Additional studies of clay deposits as alternatives to salt are warranted.

Since salt deposits are not present in all countries that are producing or that may produce nuclear wastes (*17*), some of these concerns would be alleviated if the actinides were fully recovered and recycled.

There have been other proposals for



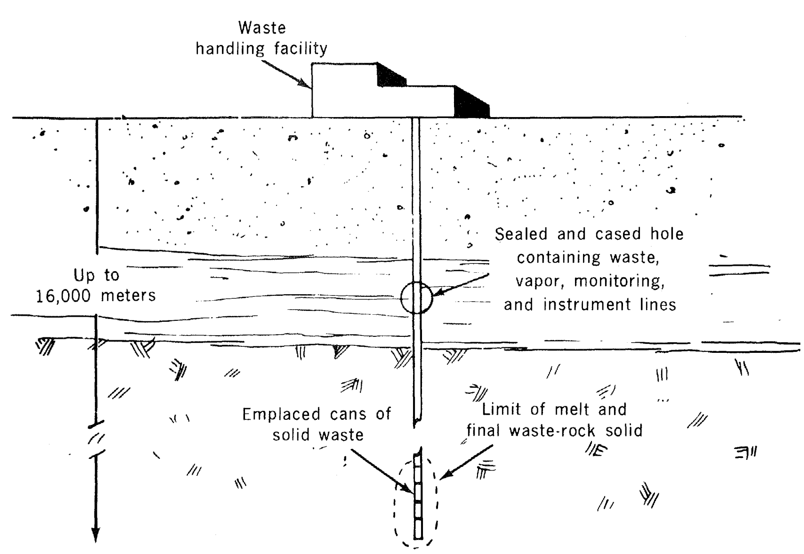


Fig. 4 (left). Solid waste emplacement in a matrix of drilled holes with no melting. [From Schneider and Platt (*9*)] Fig. 5 (right). Solid waste emplacement in a deep hole with in-place conversion to a rock waste matrix. [From Schneider and Platt (*9*)] handling high-level radioactive wastes. Three of these deserve closer attention. They are (i) seabed disposal, (ii) burial in Antarctica—in the ice sheets or more preferably subsurface burial in the ice-free areas of Antarctica, and (iii) burial in an arid zone. The first two proposals would provide the advantage of geographical isolation of the wastes from man's environment, but there are also some major problems.

Seabed Disposal

Burial in the seabed is a relative newcomer for consideration as a method of disposing of radioactive wastes. This method has been described by Bishop and Hollister (*18*) and Hollister *et al.* (*19*), but certain questions remain.

1) What is the long-term stability of the deep-sea environment for high-level radioactive wastes?

2) What would be the reaction of a water-saturated medium—such as deep-sea sediments—to long-term radiation and thermal exposure, and what would be the effects of radiolysis on the containment matrix?

3) By what processes (if any) could radionuclides migrate back to the ocean water—where it is known that they would spread rapidly at rates as high as tens of centimeters per second (*7*) and thereby cause extensive input of radionuclides to the marine environment? Dyer (*20*) has shown that migration of ^{238}Pu, ^{239}Pu, and ^{240}Pu in marine sediments near the Pacific Farollons subsites has occurred. This discovery underscores the need to understand release and transport events in the deep ocean by actual on-site measurements before placement of radioactive waste into the ocean or beneath the sediments.

4) How really stable are the midplate gyre regions?

5) What assurance do we have that deep-sea disposal will indeed isolate the wastes from man? Leakage of high-level radioactive wastes to the water column would make retrievability impossible.

6) What are the pathways via benthic fauna, the water column, and in the "boundary layer" (between sediments and ocean water) of selected radionuclides? What are the transport rates of radionuclides in these environments?

7) What are the effects of thermal and radiological regimes on the migration rate of interstitial and contained water of the sediments and subbasement of any radionuclides migrating through these subsea environments?

8) How does one implace cannisters in thousands of meters of seawater and effectively seal the hole? How does one control the free fall of any cannister and assure its "deep penetration" into the marine sediments?

These points are all extremely critical. In many marine sediments, conditions with low thermal conductivity exist. Under these conditions, very high thermal gradients would be produced in the sediments enclosing the waste cannister (the heat producer). Mass fluidization of the sediment by heat would be the result. As Hollister *et al.* (*19*) point out, "Stresses will be set up within the sediment structure by differential thermal expansion, by severe modification of the sediment/pore-water system, and possibly by phase changes. If these stresses exceed the sheer strength of the sediment, *the sediment might flow and massive convection be set up*" (italics mine). The concern here is a factor in all eight of the questions raised.

Knowledge of the detailed reactions to be expected at the various boundary layers is imperfectly understood. The obvious needs emphasis. Namely, the sea bottom is a special chemical environment, one that is very imperfectly known at this time.

In brief, undersea disposal of radioactive wastes appears, at best, to be an extremely risky scheme, one with far more potential dangers to man than land-based disposal. Considering the rapid dispersal rate and irretrievability of such wastes if dispersed in the ocean, this concept has much against it.

One last concern is that of the political ramifications of the so-called London Convention—the "Treaty for Prevention of Marine Pollution by Dumping of Wastes and Other Matter into the Ocean." This treaty addresses the issue of defining high-level radioactive wastes or other radioactive matter unsuitable for dumping at sea and indicates that wastes should be emplaced only if they have a relatively low radioactive level (*21*).

Burial in the Ice Sheet or Ice-Free Areas of Antarctica

The feasibilities of Antarctic disposal of high-level radioactive wastes have been discussed (*16, 22*). A major problem with this form of disposal would be the long-term stability of the Antarctic ice sheets. The long-term stability of the continental ice sheet of East Antarctica has been questioned by Budd and McInnes (*23*). Lack of detailed knowledge of the thermal, chemical, physical, and mechanical properties of large ice sheets is a critical factor. A different problem, that of permafrost, would arise if subsurface burial in an ice-free area were considered. The effects of thermal and radiological regimes on the permafrost of any potential ice-free burial area have to be fully evaluated, particular attention being paid to the possibility of being generated by the thermal energy available to the environment.

Waste Storage in an Arid Zone

An interesting proposition has been made by Winograd (*24*). He proposes burial of high-level radioactive wastes in the large mesas that are commonly present in arid climates, especially in the basin and range areas of the western United States. The use of arid regions for burial of certain radioactive wastes might also be possible for much longer periods of time if these areas were also tectonically stable; for example, the interior of Australia and parts of interior Asia might be suitable. If complete recycling of the actinide wastes is accomplished and only the short-lived wastes need be stored for approximately 1000 years, then this concept has much to recommend it. Winograd suggested that arid areas might be safe for waste storage for tens of thousands of years. Water would not seem to be a problem to such burial sites, since they would be located high above the valley floors of the many basins scattered throughout the arid regions of the southwestern United States. To my knowledge, this concept has not been seriously considered for short-term burial (1000 to 10,000 years) and it deserves much greater consideration than it has received.

Conclusions

Long-lived, high-level radioactive wastes require isolation and safe burial for time periods of up to 1 million years. If complete recycling and reburning of the actinide wastes is accomplished, then the wastes will require isolation for only 1000 years.

If we assume that burial in geologic deposits is the preferred method of waste disposal, then of great concern should be the question of whether we properly understand and can cope with all of the possible reactions that might develop between buried waste material and the geologic medium enclosing it at any given burial site. Clearly, the wastes should be buried in the solid form, preferably contained in an extremely stable, insoluble glass-like material. In addition, the wastes once buried should be retrievable.

If one considers the problems of seismic stability and worldwide distribution and availability of salt deposits, the overwhelming need for an international solution to the problem of the disposal of high-level radioactive wastes seems obvious.

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Posttranslational Covalent Modification of Proteins

Only 20 amino acids are used in protein synthesis, yet some 140 "amino acids" are found in various proteins.

Rosa Uy and Finn Wold

The main features of the process by which amino acids are assembled into polypeptide chains in a sequence determined by the nucleotide sequence of the genes are now well established. The process, as outlined in Table 1, is generally designated the "translation" process to emphasize that this is where the polynucleotide "language" of DNA, transcribed in messenger RNA (mRNA) molecules, is translated into the polyamino acid language of the proteins. In a strict sense, the actual translation takes place in step 1, in which each of the 20 primary amino acids is covalently attached to a specific transfer RNA (tRNA) molecule. In step 2, each of the tRNA moieties of the resulting aminoacyl-tRNA complexes is matched uniquely to a given three-nucleotide codon on the mRNA, which is bound to the synthetic machinery of the ribosomes, and the amino acid moieties are thus aligned and polymerized in the appropriate predetermined sequence. At the "full stop" codon of the mRNA, the completed polypeptide chain is released from the ribosome-mRNA complex (step 3), and the resulting linear amino acid polymer finally undergoes a number of modifications as outlined in step 4. Most current discussions of protein synthesis rather surprisingly do not include step 4. It is our thesis that since the actual gene product, the one that is most readily isolated, and the one we generally wish to characterize, is the active protein in its proper compartment of action, step 4 must be included as an essential component of the complete process of protein synthesis. Indeed, if the protein in question is subject to regulatory modulation, the form of the gene product isolated will be influenced by the modulation state (step 4b) which existed at the time of isolation. Thus, in order to understand the complete process of protein synthesis, we must understand all aspects of the processing step along with the translation and polymerization steps.

As indicated in Table 1, it is convenient to consider three distinct kinds of processing: one kind involves weak, noncovalent interactions which lead to the folding of the polypeptide chain and the association of individual chains with each other and with noncovalently bound ligands, and in turn determine the proper three-dimensional conformation of the final product. Another distinct processing step is the transport of the protein from the site of synthesis to its site of action. Considering the fact that this site of action may be an extracellular compartment or a specific intracellular organelle, it is clear that the protein may have to be transported across membranes and substantial cytoplasmic distances. Furthermore, since proteins destined for very different compartments are probably synthesized at the same site, the transport system must require a sophisticated set of traffic-directing in-

The authors are members of the Department of Biochemistry, College of Biological Sciences, University of Minnesota, St. Paul 55108.