# Radioactive waste: The problem and its management

K. R. Rao

Radioactive waste, arising from civilian nuclear activities as well as from defence-related nuclear-weapon activities, poses a formidable problem for handling and protecting the environment to be safe to the present and future generations. This article deals with this global problem in its varied aspects and discusses the cause for concern, the magnitude of the waste involved and various solutions proposed and being practised. As nuclear power and arsenal grow, continuous monitoring and immobilization of the waste over several decades and centuries and deposition in safe repositories, assumes great relevance and importance.

It's very clear
Plutonium is here to stay
Not for a year
Forever and a Day.
In time the Rockies may tumble
Yucca may crumble
They're only made of clay
But Plutonium is here to stay.

— Anonymous

'The stuff we are dealing with can't go away until it decays. You can containerize it, solidify it, immobilize it and move it, but you can't make it go away'.

– James D. Werner, Scientific American, May 1996

BEGINNING with the Manhattan Project, during the World War II, USA created a vast arsenal of nuclear weapons based on plutonium. The inputs came from a number of nuclear complexes spread across the country and they included a number of nuclear reactors to produce plutonium, reprocessing plants to extract plutonium and weapon-research laboratories and production plants. As an example, at Hanford (Washington State), a typical nuclear weapons' complex, there were 9 nuclear reactors producing plutonium, 5 reprocessing plants and 200 tanks storing nearly 200,000 m<sup>3</sup> of high-level radioactive waste.

Nearly a thousand weapons were detonated by USA for testing and the arsenal comprised of tens of thousands of weapons. The leftovers from this cold war legacy are believed to contain several large highly-contaminated reprocessing plants, thousands of tons of irradiated fuel in basins that act as 'radioactive dust-

bins', hundreds of underground tanks each containing hundreds of thousands of cubic metres of high-level radioactive waste in hazardous state, dozens of tons of unsecured plutonium and so on.

Reports from the European press state that the erstwhile Soviet Union secretly dumped nuclear reactors and radioactive waste into the bordering seas, indicating more damaging nuclear legacy of the Cold War than previously known. It is said that nuclear reactors from at least 18 nuclear submarines and icebreakers were dumped in the Barents Sea. The Russians are reported to have dumped unprocessed nuclear waste into The Sea of Japan. The latest in this scenario is that on 12 August 2000, the giant Russian nuclear submarine *Kursk*, carrying a crew of 118, sank in the icy waters of the Barents Sea after what Russian officials described as a 'catastrophe that developed at lightning speed'.

It may not be wrong to guess that any other weaponproducing complex in any other country also operates in a similar manner. Only the scale of operation may be large or small depending on the resources that are pumped in. The secrecy, callousness in handling the radioactive waste and the problems that each nation faces would be qualitatively no different; quantitatively they increase as weaponization takes deeper roots.

## Radioactive waste

Two basic nuclear reactions, namely fission of nuclei like <sup>235</sup>U, <sup>239</sup>Pu and fusion of elements like hydrogen result in release of enormous energy and radioactive elements. Controlled vast releases of energy are possible in nuclear power plant reactors through the fission reaction. The dream of controlled vast releases of energy through fusion reaction is still to be realized. Un-

K. R. Rao lives at 'Gokula', 29/2, 11th Cross Road, III Main (Margosa) Road, Malleswaram, Bangalore 560 003, India e-mail: krrao@ias.ernet.in

controlled vast releases of energy through both these reactions have been possible in 'atom' and 'hydrogen' (thermonuclear) bombs. As in many other industrial processes, in the nuclear industry also, one gets unusable and unwanted *waste* products; the residues turn out to be hazardous.

Waste, by definition, is any material (solid materials such as process residues as well as liquid and gaseous effluents) that has been or will be discarded *as being of no further use*. Note that what may be considered as one's waste may turn out to be another's wealth. Reusable plastics and other components in day-to-day household waste are good examples in this context. This concept holds good for radioactive waste also, in some sense. Waste that emits nuclear radiation is radioactive waste. (See Box 1 for basic concepts of radioactivity.)

#### Natural radioactivity

It is somewhat surprising that nature has been a large producer of radioactive waste. Over the eons, the surface of the Earth and the terrestrial crust happens to be an enormous reservoir of primordial radioactivity. Small amounts of radioactive materials are contained in mineral springs, sand mounds and volcanic eruptions. Essentially all substances contain radioactive elements of natural origin to some extent or the other.

The second source of radioactive waste is a part of industrial mining activity where, during mineral exploration and exploitation, one excavates the primordial material from the Earth that contains radioactivity, uses part of it and rejects the radioactive residues as waste. These are referred to as Naturally Occurring Radioactive Materials (NORMs) and are ubiquitous as residual wastes in processing industries that cover fertilizers, iron and steel, fossil fuel, cement, mineral sands, titanium, thorium and uranium mining as well as emanations and waste from coal and gas-fired power plants.

One should note that in many industries, radiation exposure to the workers and the general public would be at least as high as those from nuclear installations and in some cases it is even higher. It is also known that certain mineral springs contain fairly large amounts of <sup>222</sup>radon. Monazite sand deposits in coastal areas may result in radiation exposure to humans around an order of magnitude in excess of the currently set international exposure limits to radioactive waste disposal (one msv/ year) and volcanic deposits result in similar exposure. There is no place on Earth that is free from natural radioactive background; it may vary from place to place all the way from the low to the high. The content of

## Box 1. Radioactivity

Certain elements that compose matter emit particles and radiations spontaneously. This phenomenon is referred to as 'radioactivity', it cannot be altered by application of heat, electricity or any other force and remains unchangeable.

Three different kinds of rays, known as alpha, beta and gamma rays are associated with radioactivity. The alpha rays consist of particles (nuclei of helium atoms) carrying a positive charge, beta rays particles have negative charge (streams of electrons) and gamma rays are chargeless electromagnetic radiation with shorter wavelengths than any X-rays. These 'rays' can penetrate living tissues for short distances and affect the tissue cells. But because they can disrupt chemical bonds in the molecules of important chemicals within the cells, they help in treating cancers and other diseases. Every element can be made to emit such rays artificially. If such radioactive elements are placed in the body through food or by other methods, the rays can be traced through the body. This use of tracer elements is extremely helpful in monitoring life processes. Geologists use radioactivity to determine the age of rocks. As atoms lose particles as heavy as nuclei of helium, they become atoms of some other element. That is, the elements change or 'transmute' into other elements until the series ends with a stable element.

Radioactive elements decay at different rates. Rates are measured as half-lives – that is, the time it takes for one half of any given quantity of a radioactive element to disintegrate. The longest half-life is that of the 'isotope' <sup>238</sup>U of uranium. It is 4.5 billion years. Some isotopes have half-lives of years, months, days, minutes, seconds, or even less than millionths of a second.

Measurement units and permissible dosages

Radioactivity is measured in Becquerel (Bq) units. 1 Bq = 1 decay or disintegration per second. Curie (Ci) was used earlier and 1 Ci = 37 billion Bq  $(3.7 \times 10^{10} \text{ disintegrations per second})$  or 37 Bq = 1 nano-Ci.

To measure the health risk through ionization, in the US the most commonly used unit is rem or mrem (millirem). In Europe, the most commonly used measuring unit for this purpose is Sv (Sievert) or mSv (milli-Sv). Conversion of rem to Sieverts: 1 rem = 0.01 Sv = 10 mSv.

radioactivity in the seas is estimated to be nearly 10,000 exabecquerel (Ebq =  $10^{18}$  Bq). The residual waste tailings from past mining and milling operations are estimated to be around several million tons at many places and the radioactivity contained may be nearly 0.001 EBq. Thousands of such sites are scattered all around the world.

At OKLO, located in Gabon in the West African rainforest, there exists uranium ore that formed an active natural reactor over some billion years ago. A study of this site has shown that the actinide fission products migrated, under highly unfavourable conditions, only a few tens of metres during this long duration.

#### Artificial radioactivity

Radioactivity was discovered about a hundred years ago. Following the Second World War and discovery of the fission process, human activity added radioactivity artificially to the natural one. Two main sources have been: (a) the civilian nuclear programmes, including nuclear power production, medical and industrial applications of radioactive nuclides for peaceful purposes, and (b) the military nuclear programme, including atmospheric and underground nuclear-weapon testing and weapon production (see Box 2 for the nature of artificial radioactive isotopes produced).

#### Nuclear fuel cycle

As stated earlier, civilian nuclear operations lead to radioactivity. The story of uranium from its mining to its use in reactors and thence of chemical processing and accumulation of radioactive waste is covered by what is referred to as 'nuclear fuel cycle' (see Box 3 for a schematic fuel cycle). The ore that is mined in uranium mines is sent to a uranium mill, where a small uraniumcontaining fraction is separated from the ore, leaving behind virtually almost the entire ore in the tailings. The uranium fraction is processed to recover pure uranium in metallic form. Uranium metal consists of the isotope <sup>235</sup>U to the extent of 0.7%, the remaining 99.3% being <sup>238</sup>U. <sup>235</sup>U fissions on absorption of thermal neutrons, while <sup>238</sup>U does not. Hence this small fraction of <sup>235</sup>U is 'enriched' for use in light-water reactors for deriving power. Highly enriched <sup>235</sup>U is used for nuclear weapons also. In CANDU-type heavy-water reactors, one can use natural uranium itself as fuel, without any enrichment. (Except for the reactors at Tarapore and the fast-breeder test reactor at Kalpakkam, the other Indian power and research reactors use natural uranium as fuel.) Fresh fuel made of uranium (sometimes containing plutonium, in addition) is weakly radioactive. The fuel, after sufficient use in reactors, is referred as 'spent fuel'; the 'ash' after 'burning' the fuel contains fissionfragment debris from spontaneous or neutron-induced

Isotope	Half-life	Isotope	Half-life	Isotope	Half-life
Relatively short hai	f-life				
Strontium-89	54 days	Zirconium-95	65 days	Niobium-95	39 days
Ruthenium-103	40 days	Rhodium-103	57 minutes	Rhodium-106	30 seconds
lodine-131	8 days	Xenon-133	8 days	Tellurium-134	42 minutes
Barium-140	13 days	Lanthanum-140	40 h	Cerium-141	32 days
Year to century-sca	ale half-life*				
Hydrogen-3	12 years	Krypton-85	10 years	Strontium-90	29 years
Ruthenium-106	1 year	Cesium-137	30 years	Cerium-144	1.3 years
Promethium-147	2.3 years	Plutonium-238	85.3 years	Americium-241	440 years
Curium-224	17.4 years				
Longer half-life					
Technecium-99 Plutonium-240	2 × 10 <sup>6</sup> years 6500 years	lodine-129 Americium-243	$1.7 \times 10^7$ years 7300 years	Plutonium-239	24000 years

<sup>\*</sup>Half-lives of the order of years to decades of isotopes of elements that can seek tissues or organs biologically (being akin to other elements chemically) are the most hazardous from point of view of radiation. For example, <sup>90</sup>Sr, being chemically akin to Ca, can seek the bone and lodge itself there for years causing radioactive damage to surrounding tissues.

Nuclear fuel cycle: The activities comprising mining, processing, fuel fabrication and ultimately use of fuel in nuclear reactors result in power generation. Reprocessing spent fuel helps in recycling plutonium for fuel fabrication. The byproducts in this activity are enriched fissile material useful for fuel as well as for weapons, depleted uranium used for DU shells, the actinides and radioactive waste. Processing Uranium mining Uranium Uranium plant Waste DU Shells for Tanks Depleted uranium Enrichment plant Fuel Nuclear reactor <sup>235</sup>U Weapons From uranium mining to nuclear reactor Thorium sands Thorium Irradiated thorium Processing Nuclear reactor Nuclear reactor Reprocessing Weapon Waste From thorium mining to nuclear reactor Nuclear reactor Spent fuel Plutonium Reprocessing plan Fuel High level Nuclear reactor Weapons Nuclear power radioactive residue Actinides Processing Waste Nuclear reactor to nuclear power, spent fuel and reprocessing

fission of uranium and actinides, actinide elements and unutilized uranium. This irradiated fuel is highly radio-active. Transuranic actinides (principally neptunium, plutonium, americium and curium) are created by absorption of neutrons in non-fissioned uranium and by sequential absorption of neutrons in the consequently

formed daughter elements. Although nearly 200 radionuclides are produced during the burn-up of the fuel, the great majority of them are relatively short-lived and decay to low levels within a few decades (see Box 2). Hence the spent fuel is often allowed to 'cool' in spentfuel bays of water, to allow short-lived radioactivity to

1537

decay. Often such fuel is stored for indefinite time in the fuel pools without any further processing, or in dry 'coffins'. The short-lived radionuclides therefore do not pose a big problem for long-term disposal.

The spent fuel, when subjected to chemical processing, yields uranium and plutonium fractions apart from the rest of the 'ash'. In this article we do not deal with the chemistry involving a variety of highly toxic chemicals or the complex chemical processes that one employs, either in fuel reprocessing or in radioactive waste management. Beginning with dissolution of cladded burnt fuel to retrieving useful fissile elements is an enormous activity involving chemical engineering, remote handling, monitoring, etc. As opposed to the socalled 'once-through fuel cycle' wherein no material is recycled, in the 'closed-cycle fuel cycle', uranium is recycled for fuel production and plutonium for either fuel production or for weapons. Normally, the irradiated uranium is dissolved in an acid medium and treated with organic solvents to recover plutonium and remnants of uranium. The byproduct is a highly acidic liquid, a high-level radioactive waste containing fission fragments and transuranic elements. The transuranic elements can be separated further, as they constitute rare, precious and often fissile materials themselves. This is the 'wealth' from the waste we referred to in the beginning.

## The cause for concern

Radioactive waste, whether natural or artificial, is a potential harbinger of radioactive exposure to humans through many channels. The routes are direct exposure to materials that are radioactive, inhalation and ingestion of such materials through the air that one breathes or food that one consumes. The quantum of exposure (dose × duration of exposure) decides the deleterious effects that may result. Exposure may occur to particular organs locally or to the whole body. Sufficiently high exposure can lead to cancer (see Box 4). The radiotoxicity of a particular radionuclide is quantified in terms of what is referred to as 'potential hazard index' that is defined in terms of the nuclide availability, its activity, maximum permissible intake annually and its half-life. This depends on a variety of factors like physical half-life, biological half-life, sensitivity of the organ or tissue where the nuclide is likely to concentrate, ionizing power of the radiation from the nuclide that depends on the energy of the radiation emitted from the radionuclide, etc. It is from such considerations that one concludes that radioactive nuclides of elements like <sup>137</sup>Cs or <sup>90</sup>Sr or <sup>131</sup>I are the most hazardous on the scale of a human beings' lifetime. Other long-life nuclides like <sup>239</sup>Pu, <sup>241</sup>Am, <sup>237</sup>Np pose a long-term hazard, on the other hand, to future generations.

## Box 4. Radiation effects.

Every inhabitant on this planet is constantly exposed to naturally occurring ionizing radiation called background radiation. Sources of background radiation include cosmic rays from the Sun and stars, naturally occurring radioactive materials in rocks and soil, radionuclides normally incorporated into our body's tissues, and radon and its products, which we inhale. We are also exposed to ionizing radiation from man-made sources, mostly through medical procedures like X-ray diagnostics. Radiation therapy is usually targeted only to the affected tissues.

Much of our data on the effects of large doses of radiation comes from survivors of the atomic bombs dropped on Hiroshima and Nagasaki in 1945 and from other people who received large doses of radiation, usually for treatment. Only about 12% of all the cancers that have developed among those survivors are estimated to be related to radiation.

lonizing radiation can cause important changes in our cells by breaking the electron bonds that hold molecules together. For example, radiation can damage our genetic material (DNA). But the cells also have several mechanisms to repair the damage done to DNA by radiation.

Potential biological effects depend on how much and how fast a radiation dose is received. An acute radiation dose (a large dose delivered during a short period of time) may result in effects which are observable within a period of hours to weeks. A chronic dose is a relatively small amount of radiation received over a long period of time. The body is better equipped to tolerate a chronic dose than an acute dose as the cells need time to repair themselves.

Radiation effects are also classified in two other ways, namely *somatic* and *genetic* effects. Somatic effects appear in the exposed person. The delayed somatic effects have a potential for the development of cancer and cataracts. Acute somatic effects of radiation include skin burns, vomiting, hair loss, temporary sterility or subfertility in men, and blood changes. Chronic somatic effects include the development of eye cataracts and cancers. The second class of effects, namely genetic or *heritable* effects appears in the future generations of the exposed person as a result of radiation damage to the reproductive cells, but risks from genetic effects in humans are seen to be considerably smaller than the risks for somatic effects.

Although nature's sources are to be as much feared as those from artificial sources, the (atomic) bomb's legacy has set a certain perception in the public mind, of the dangers inherent or implicit in the use and abuse of nuclear facilities, operations and waste. The recent emphasis arises because of concern to the effects on the environment over a very long period of time. High-level radioactive waste is potentially toxic for tens of thousands to millions of years; it is also the most difficult to be disposed safely because of its heat and radiation output. Thermal, chemical and radiological gradients operate on the environment over periods as long as 500,000 years.

Some of the concerns being expressed border on over-reaction to a problem that exists. It is not that one should wish away the problem. But on the other hand, the reaction or concern is often inflated. As Tanner asked 'Are not we kidding ourselves when we claim to be so concerned about the far-out possibility that a nuclear-waste-disposal site may begin to leak 10,000 or 1,000,000 years from now? In what other area of life do we show such foresight?' (*Phys. Today*, January 1998, p. 86.)

We are confronted with a dilemma. On one side, 50–100 years hence, our fossil fuel sources may be reaching the rock-bottom of availability and the renewable sources of energy (solar, wind, geothermal, etc. power sources) may not meet the demands of society. Till alternate energy sources are developed, the only source available to mankind is the nuclear power.

To set the scenario in proper perspective, it should be noted that nuclear power plants are managed subject to several radiation protection control practices. Secondly, one may also note that 'a 1000 MW electric coal-fired power plant releases into the environment nearly 6 million tonnes of greenhouse gases, 500,000 tons of mixtures of sulphur and nitrogen oxides and about 320,000 tonnes of ashes'. These ashes containing NORMs are potentially capable of subjecting humanity to a collective dose of radiation higher than that attributable to wastes discharged into the environment by nuclear power plants generating the same amount of electricity. In spite of this ground reality, public perception about nuclear wastes is rather skewed against nuclear power in several countries.

#### Quantifying natural and artificial nuclear waste

The level of radioactive waste is quoted in terms of volume (in cubic metres) or in tonnage. Another way is to quote the radioactivity contained in such waste in bequerels (Bq). Both the units are useful because one needs to know the volume or weight of the waste to be handled for disposal purposes and also the radioactivity contained therein.

We have already noted that nuclear waste from natural sources, including mining and related operations, could have resulted in production of radioactive waste of a few EBq and the sea is repository of several thousand EBq of radioactivity.

Compared to this it is estimated that in the military nuclear operations, the cold-war era resulted in release of more than 1000 EBq of nuclear debris in the atmosphere. Production of weapon-grade material resulted in about 1000 EBq of residual waste and 'accidents and losses' of nuclear submarines and nuclear-powered satellites might have resulted in waste of a few EBq.

In the civilian regime, it is estimated that the nuclear waste, as a result of nuclear power production around the world over the past 50 years, is of the order of 1000 EBq and is growing at the rate of approximately 100 EBq/year. Typically, a large nuclear power plant of generating capacity of 1000 MW electricity produces 'around 27 tonnes of high-level radioactive waste, 310 tonnes of intermediate-level and 460 tonnes of low-level radioactive waste'.

#### Classification of radioactive waste

Nuclear waste can be generally classified as either 'lowlevel' radioactive waste or 'high-level' radioactive waste.

#### Low-level radioactive waste

Basically all radioactive waste that is not high-level radioactive waste or intermediate-level waste or transuranic waste is classified as low-level radioactive waste. Volume-wise it may be larger than that of highlevel radioactive waste or intermediate-level radioactive waste or transuranic waste, but the radioactivity contained in the low-level radioactive waste is significantly less and made up of isotopes having much shorter halflives than most of the isotopes in high-level radioactive waste or intermediate-level waste or transuranic waste. Large amounts of waste contaminated with small amounts of radionuclides, such as contaminated equipment (glove boxes, air filters, shielding materials and laboratory equipment) protective clothing, cleaning rags, etc. constitute low-level radioactive waste. Even components of decommissioned reactors may come under this category (after part decontamination procedures).

The level of radioactivity and half-lives of radioactive isotopes in low-level waste are relatively small. Storing the waste for a period of 10 to 50 years will allow most of the radioactive isotopes in low-level waste to decay, at which point the waste can be disposed of as normal refuse.

It may come as a surprise that several investigations have shown that exposure of mammals to low levels of radiation may indeed be beneficial, including, 'increased life span, greater reproductive capacity, better disease resistance, increased growth rate, greater resistance to higher radiation doses, better neurological function, better wound healing and lower tumour induction and growth' (Devaney, J. J., *Phys. Today*, January 1998, p. 87). Beneficial effects on plants include accelerated growth and development and increased harvests. Low-level radioactive waste, therefore, seems to be benign.

#### High-level radioactive waste

High-level radioactive waste is conceptualized as the waste consisting of the spent fuel, the liquid effluents arising from the reprocessing of spent fuel and the solids into which the liquid waste is converted. It consists, generally, material from the core of a nuclear reactor or a nuclear weapon. This waste includes uranium, plutonium and other highly radioactive elements created during fission, made up of fission fragments and transuranics. (Note that this definition does not specify the radioactivity that must be present to categorize as high-level radioactive waste.) These two components have different times to decay. The radioactive fission fragments decay to different stable elements via different nuclear reaction chains involving  $\alpha$ ,  $\beta$  and  $\gamma$  emissions to innocuous levels of radioactivity, and this would take about 1000 years. On the other hand, transuranics take nearly 500,000 years to reach such levels. Heat output lasts over 200 years. Most of the radioactive isotopes in high-level waste emit large amounts of radiation and have extremely long half-lives (some longer than 100,000 years), creating long time-periods before the waste will settle to safe levels of radioactivitv.

As a thumb-rule one may note that 'volumes of low-level radioactive waste and intermediate-level waste greatly exceed those of spent fuel or high-level radioactive waste'. In spite of this ground reality, the public concerns regarding disposal of high-level radioactive waste is worldwide and quite controversial.

### Approaches to radioactive waste disposal

Waste disposal is discarding waste with no intention of retrieval. Waste management means the entire sequence of operations starting with generation of waste and ending with disposal.

Solid waste disposal, of waste such as municipal garbage, is based on three well-known methods, namely landfills, incineration and recycling. Sophisticated methods of landfills are adapted for radioactive waste also. However, during incineration of ordinary waste, fly ash, noxious gases and chemical contaminants are released into the air. If radioactive waste is treated in this manner, the emissions would contain radioactive particulate matter. Hence when adapted, one uses fine particulate filters and the gaseous effluents are diluted and released. Recycling to some extent is feasible. We have already dealt with the reprocessing approach, whereby useful radioactive elements are recovered for cyclic use. But it still leaves some waste that is a part of the high-level radioactive waste.

Radioactive waste management involves minimizing radioactive residues, handling waste-packing safely, storage and safe disposal in addition to keeping sites of origin of radioactivity clean. Poor practices lead to future problems. Hence choice of sites where radioactivity is to be managed safely is equally important in addition to technical expertise and finance, to result in safe and environmentally sound solutions.

The International Atomic Energy Agency (IAEA) is promoting acceptance of some basic tenets by all countries for radioactive waste management. These include: (i) securing acceptable level of protection of human health; (ii) provision of an acceptable level of protection of environment; (iii) while envisaging (i) and (ii), assurance of negligible effects beyond national boundaries; (iv) acceptable impact on future generations; and (v) no undue burden on future generations. There are other legal, control, generation, safety and management aspects also.

Next we review some approaches for radioactive waste disposal.

To begin with, the radioactive waste management approach is to consider the nature of radioactive elements involved in terms of their half-lives and then choose the appropriate method of handling. If the concentrations of radioactive elements are largely shortlived, then one would resort to what is referred to as 'delay and decay' approach; that is, to hold on to such a waste for a sufficiently long time that the radioactivity will die in the meanwhile. A second approach is to 'dilute and disperse' so that the hazard in the environment is minimized. But when the radioactivity is long-lived, the only approach that is possible is to 'concentrate and contain' the activity. In order to carry out concentrating the waste (generally the sludge), chemical precipitation, ion exchange, reverse osmosis and natural or steam evaporation, centrifuging, etc. are resorted to. The resulting solids are highly concentrated in radioactivity. In the following we shall discuss some of the approaches that are being advocated or are currently in practice.

However, to the extent that the mining operations result in 'bringing the radioactivity to the surface and change its chemical and physical form that may increase its mobility in the environment', they assume importance in radioactive waste management. Long-lived isotopes like <sup>230</sup>Th, <sup>226</sup>Ra, the decay products of uranium are part of the tailings and hence the tailings have to be contained.

Low-level radioactive waste and even transuranic waste is often buried in shallow landfills. One has to pay attention to any groundwater contamination that may result due to this.

The highly radioactive liquid effluents are expected to be ultimately solidified into a leach-resistant form such as borosilicate glass, which is fairly robust in the sense that it is chemically durable, resistant to radiolysis, relatively insensitive to fluctuations in waste composition and easy to process remotely. (Immobilization in cement matrices or bitumanization or polymerization are also some of the other options that are practised to some extent.) However, it must be noted that plutonium does not bind strongly to the matrix of the glass and 'thus can be loaded only in trace amounts to prevent the possibility of criticality or recovery for clandestine purposes'. This glass in turn is placed in canisters made of specific alloys. Choice of the canister material would depend on the ultimate site where the waste will be disposed-off. For example, if the ultimate disposal is in the oceans, the alloy chosen must have low corrosion rates under the environmental temperature, pressure, oxygen concentration, etc. Studies have been carried out in this respect. For example, it is found that in oxygenated sea water at 250°C, 7 mega Pascals pressure and 1750 ppm of dissolved oxygen, the corrosion rates of 1018 mild steel, copper, lead, 50:10 cupro-nickel, Inconel 600 and Ticode 12 are 11.0, 5.0, 1.0, 0.7, 0.1 and 0.06 mm/year, respectively.

One seeks to dispose-off the high-level radioactive waste packages contained in multiple metal-barrier canisters within natural or man-made barriers, to contain radioactivity for periods as long as 10,000 to 100,000 years. 'The barrier is a mechanism or medium by which the movement of emplaced radioactive materials is stopped or retarded significantly or access to the radioactive materials is restricted or prevented'. It is obvious that recourse to multiple barriers may assure safety of emplaced radioactivity over long periods of time. The man-made barriers, namely the form to which waste is reduced, for example, in the glassy form, and the canister along with overpackaging, go along with natural barriers. As far as the choice of natural barriers is concerned, land-based mined depositories over fairly stable geologic formations are preferred over disposal in the oceans. However several social and environmental concerns have prevented the land-route being adopted in counties like USA even after 50 years of accumulation of radioactive waste. Therefore proposals have been made to take to the ocean-route and there also the choice varies from just placement of the canisters over the seabed to placement within the sub-seabed sediments and even within the basement rocks.

In the US, as spent fuels have reached levels of radioactivity of the order of 50,000 MCi (excluding military sources), there is dearth of space to store additional irradiated fuel removed from operating reactors. Legally, the Department of Energy (DOE) is expected to take charge of all commercial spent fuel. However, the DOE has run into a dead-end. On one hand it is unable to use spent fuel and on the other, its attempts to develop a permanent repository at Yucca Mountain in Nevada are met by social and State challenges as well as lack of complete study of the site itself. Presidential consent has not been forthcoming to any legislation in this connection.

#### Options being aired for disposing radioactivity

Triet Nguyen, Department of Nuclear Engineering, University of California, Berkeley, has written in an article 'High-level Nuclear Waste Disposal', 14 November 1994 that 'High-level nuclear waste from both commercial reactors and defence industry presents a difficult problem to the scientific community as well as the public. The solutions to this problem are still debatable, both technically and ethically .... There are many proposals for disposing high-level nuclear wastes. However the most favoured solution for the disposal of these wastes is isolating radioactive waste from man and biosphere for a period of time such that any possible subsequent release of radionuclides from the waste repository will not result in undue radiation exposure. The basic idea behind this is to use stable geological environments that have retained their integrity for millions of years to provide a suitable isolation capacity for the long time-periods required. The reason for relying on such geological environments is based on the following main consideration: 'Geological media is an entirely passive disposal system with no requirement for continuing human involvement for its safety. It can be abandoned after closure with no need for continuing surveillance or monitoring. ... The safety of the system is based on multiple barriers, both engineered and natural, the main one being the geological barrier itself.' One way of disposing high-level nuclear waste materials which meets the above condition is the concept of disposing of these wastes by burial in suitable geologic media beneath the deep ocean floor, which is called seabed disposal.

The following options have been aired sometime or the other. Each one of the options demands serious studies and technical assessments:

- Deep geological repositories
- Ocean dumping

- Seabed burial
- Sub-seabed disposal
- Subductive waste disposal method
- Transforming radioactive waste to non-radioactive stable waste
- Dispatching to the Sun.

Major problems due to legal, social, political and financial reasons have arisen in execution due to

- Environmental perceptions
- · Lack of awareness and education
- 'Not-in-my-backyard' syndrome
- 'Not-in-the-ocean' syndrome
- Lack of proven technology.

#### Geologic disposal

Geologic disposal in deep geological formations — whether under continental crust or under seabed — as a means of radioactive waste disposal has been recognized since 1957, for handling long-lived waste. Quite often, contrary to views expressed by environmentalists, it is 'not chosen as a cheap and dirty option to get the radioactive waste simply "out of site and out of mind".

The deep geological sites provide a natural isolation system that is stable over hundreds of thousands of years to contain long-lived radioactive waste. In practice it is noted that low-level radioactive waste is generally disposed in near-surface facilities or old mines. High-level radioactive waste is disposed in host rocks that are crystalline (granitic, gneiss) or argillaceous (clays) or salty or tuff. Since, in most of the countries, there is not a big backlog of high-level radioactive waste urgently awaiting disposal, interim storage facilities, which allow cooling of the wastes over a few decades, are in place.

#### Ocean-dumping

For many years the industrialized countries of the world (e.g. USA, France, Great Britain, etc.) opted for the least expensive method for disposal of the wastes by dumping them into the oceans. Before 1982, when the United States Senate declared a moratorium on the dumping of radioactive wastes, the US dumped an estimated 112,000 drums at thirty different sites in the Atlantic and Pacific oceans.

Though this practice has been banned by most of the countries with nuclear programmes, the problem still persists. Russia, which currently controls sixty per cent of the world's nuclear reactors, continues to dispose of its nuclear wastes into the oceans. According to Russia's Minister of Ecology, it will continue to dump its wastes into the oceans because it has no other alterna-

tive method. It will continue to do so until it receives enough international aid to create proper storage facilities. In response, the United States has pledged money to help Russia, but the problem continues.

Although radioactive waste has known negative effects on humans and other animals, no substantial scientific proof of bad effects on the ocean and marine life has been found. Hence some nations have argued that ocean-dumping should be continued. Others argue that the practice should be banned until further proof of no harm is available.

Oceanic Disposal Management Inc., a British Virgin Islands company, has also proposed disposing of nuclear and asbestos waste by means of Free-Fall Penetrators. Essentially, waste-filled missiles, which when dropped through 4000 m of water, will embed themselves 60–80 m into the seabed's clay sediments. These penetrators are expected to survive for 700 to 1500 years. Thereafter the waste will diffuse through the sediments. This was a method considered by the Scientific Working Group (SWG) of the Nuclear Energy Agency (NEA) during the eighties.

Penetrator disposal is potentially both feasible and safe, its implementation would depend on international acceptance and the development of an appropriate international regulatory framework. Neither of these exists, nor are they likely to in the foreseeable future. The penetrator method has also been further constrained by a recent revision of the definition of 'dumping', by the London Dumping Convention, to include 'any deliberate disposal or storage of wastes or other matter in the seabed and the subsoil thereof'.

#### Sub-seabed disposal

Seabed disposal is different from sea-dumping which does not involve isolation of low-level radioactive waste within a geological strata. The floor of deep oceans is a part of a large tectonic plate situated some 5 km below the sea surface, covered by hundreds of metres of thick sedimentary soft clay. These regions are desert-like, supporting virtually no life. The Seabed Burial Proposal envisages drilling these 'mud-flats' to depths of the order of hundreds of metres, such boreholes being spaced apart several hundreds of metres. The high-level radioactive waste contained in canisters, to which we have referred to earlier, would be lowered into these holes and stacked vertically one above the other interspersed by 20 m or more of mud pumped in.

The proposal to use basement-rock in oceans for radioactive waste disposal is met with some problems: variability of the rock and high local permeability. Oceanic water has a mixing time of the order of a few thousand years which does not serve as a good barrier for long-lived radionuclides.

Since experiments cannot be conducted to assure safety of seabed disposal on the basis of actual canisters deposited in the seabed over periods of interest, namely over hundreds of thousands of years, model calculations have been performed to predict the capabilities of such a disposal option.

The model approach has started with selection of sites and acquisition of site-specific data using marine geological methods. These sites are away from deep-sea trenches, mid-oceanic ridges or formation zones where geological activities are high. These sites are also far away from biologically productive areas in the oceans. The sediments in chosen sites are fine-grained and are called 'abyssal red clay'. These sites are believed to have desirable barrier properties with 'continuous stable and depositional histories'. Therefore these potential waste repositories are geologically stable over periods of the order of  $10^7$  years and are likely not to have human activities, as they are not resources of fishes or hydrocarbons or minerals.

Core samples from most Pacific and Atlantic sites have been studied to investigate thermal, chemical and radiological effects. It is found that when sea water and sample sediment mixtures are heated at 300°C at high pressure, the solution pH changes from 8 to 3. Calculations suggest that 'less than 2 cubic metres of untreated sediment would be needed to neutralize all the acid generated in the thermally perturbed region of about 5.5 m³. The canister material has to be compatible with this type of environment for periods of at least 500 years by which time fission fragment activity would become acceptable. Similarly, other calculations have taken into account sediment–nuclide interactions to determine ion concentration around a buried source as a function of time.

Experimental work has already established that clays have the property of holding on to several radioactive elements, including plutonium; hence, seepage of these elements into saline water is minimal. Rates of migration of these elements over hundreds of thousands of years would be of the order of a few metres. Hence, during such long times, radioactivity will diminish to levels below the natural radioactivity in sea water due to natural radioactive decay. The clays also have plastic-like behaviour to form natural sealing agents. Finally, the mud-flats have rather low permeability to water; hence, leaching probability is rather low.

It may be noted that the method depends on standard deep-sea drilling techniques routinely practised and sealing of the bore-holes. These two aspects are well-developed, thanks to the petroleum industry and also because of an international programme called the Ocean Drilling Programme. Core samples from about half a dozen vastly separated sites in the Pacific and Atlantic oceans have 'showed an uninterrupted history of geological tranquillity over the past 50–100 million years'.

However there are questions that remain to be answered:

- Whether migration of radioactive elements through the ocean floor is at the same rate as that already measured in the laboratories?
- What is the effect of nuclear heat on the deep oceanic-clavs?
- What is the import on the deep oceanic fauna and waters above?
- In case the waste reaches the seabed-surface, will the soluble species (for example, Cs, Tc, etc.) be diluted to natural background levels? If so, at what rate?
- What happens to insoluble species like plutonium?
- What is the likelihood of radioactivity reaching all the way to the sea surface?
- In problems of accidents in the process of seabed burial leading to, say, sinking ships, to loss of canisters, etc. how does one recover the waste-load under such scenarios?
- What is the likelihood that the waste is hijacked from its buried location?

Added to these technical problems are others:

- International agreement to consider seabed-burial as distinct from 'ocean-dumping'.
- This method would be expensive to implement, but its cost would be an impediment to any future plutonium-mining endeavour.

Although the world trend is toward the option of land-based disposal, it is doubtful whether restricting repositories to land-based sites really helps prevention of sea pollution. If radionuclides from a land-based repository leached out to the surface, they would be quickly transported to the sea by surface water. What is essential is to isolate radionuclides from the biosphere as reliably as possible. If sub-seabed disposal results in more reliable isolation, sub-seabed disposal is the better safeguard against sea pollution. This method takes into consideration technological feasibility, protection of marine environments, and availability of international understanding.

The United Nation's Convention on the Law of the Sea delineates that a coastal state is granted sovereign rights to utilize all resources in water and under the seabed within its exclusive economic zone (EEZ), which can extend from the coast line up to 200 nautical miles (about 370 km) offshore. A repository is proposed to be constructed in bedrock 2 km beneath the seabed. To utilize sub-seabed disposal within the EEZ, it is also proposed that waste packages would be transported through a submarine tunnel connecting land with the

sub-seabed repository. Sea pollution by an accident during disposal work would be improbable, because waste would never go through sea water during the work. The proposed method is a variation of geologic disposal. Long-term monitoring is also possible by maintaining the access tunnel for some time after constructing artificial barriers.

While sub-seabed disposal of nuclear waste-filled canisters thrown from vessels apparently is regulated by the London Convention, it is not prohibited or regulated by the London Convention when accessed via land-based tunnels. Sweden has been practising this method of sub-seabed disposal since 1988, when a repository for reactor wastes was opened sixty metres below the Baltic seabed. This project has been widely cited by politicians from other countries as a great example of solving the nuclear waste problem. Because of Sweden's initiative, nuclear waste is already being deposited under the seabed. Other countries could follow Sweden's example and dispose-off nuclear waste under the seabed via land-based tunnels.

## Subductive waste disposal method

This method is the state-of-the-art in nuclear waste disposal technology. It is the single viable means of disposing radioactive waste that ensures non return of the relegated material to the biosphere. At the same time, it affords inaccessibility to eliminated weapons material. The principle involved is the removal of the material from the biosphere faster than it can return. It is considered that 'the safest, the most sensible, the most economical, the most stable long-term, the most environmentally benign, the most utterly obvious places to get rid of nuclear waste, high-level waste or low-level waste is in the deep oceans that cover 70% of the planet'.

Subduction is a process whereby one tectonic plate slides beneath another and is eventually reabsorbed into the mantle. The subductive waste disposal method forms a high-level radioactive waste repository in a subducting plate, so that the waste will be carried beneath the Earth's crust where it will be diluted and dispersed through the mantle. The rate of subduction of a plate in one of the world's slowest subduction zones is 2.1 cm annually. This is faster than the rate (1 mm annually) of diffusion of radionuclides through the turbidite sediments that would overlay a repository constructed in accordance with this method. The subducting plate is naturally predestined for consumption in the Earth's mantle. The subducting plate is constantly renewed at its originating oceanic ridge. The slow movement of the plate would seal any vertical fractures over a repository at the interface between the subducting plate and the overriding plate.

## Transmutation of high-level radioactive waste

This route of high-level radioactive waste envisages that one may use transmutational devices, consisting of a hybrid of a subcritical nuclear reactor and an accelerator of charged particles to 'destroy' radioactivity by neutrons. 'Destroy' may not be the proper word; what is effected is that the fission fragments can be transmuted by neutron capture and beta decay, to produce stable nuclides. Transmutation of actinides involves several competing processes, namely neutron-induced fission, neutron capture and radioactive decay. The large number of neutrons produced in the spallation reaction by the accelerator are used for 'destroying' the radioactive material kept in the subcritical reactor. The scheme has not yet been demonstrated to be practical and cost-effective.

#### Solar option

It is proposed that 'surplus weapons' plutonium and other highly concentrated waste might be placed in the Earth orbit and then accelerated so that waste would drop into the Sun. Although theoretically possible, it involves vast technical development and extremely high cost compared to other means of waste disposal. Robust containment would be required to ensure that no waste would be released in the event of failure of the 'space transport system'.

#### Other options and issues

In its 1994 report entitled 'Management and Disposition of Excess Weapons' Plutonium', the National Academy of Sciences set forth two standards for managing the risks associated with surplus weapons-usable fissile materials. First, the storage of weapons should not be extended indefinitely because of non-proliferation risks and the negative impact it would have on arms-reduction objectives. Second, options for long-term disposition of plutonium should seek to meet a 'spent-fuel standard' in which the plutonium is made inaccessible for weapons use.

One of the chosen options of DOE is for dealing with surplus plutonium, its use as a Mixed Oxide Fuel (MOX) to be burned in reactors such as the CANDU.

The United States policy is not to encourage the civil use of plutonium. The Nuclear Control Institute regards the vitrification approach as posing fewer risks than the MOX approach with regard to diversion or theft of warhead material, reversal of the disarmament process, and other adverse effects on international arms control and non-proliferation efforts. A decision to dispose-off warhead plutonium by means of vitrification or other immobilization technology would be an essential step

toward achievement of such a regime. Proponents of MOX disposition claim that vitrification technology is immature, speculative and cannot be ready soon enough. On the other hand, the MOX option, though it does not necessarily involve further reprocessing, would clearly encourage civilian use of plutonium, which in some countries like Japan even includes plans for reprocessing irradiated MOX fuel. In the opinion of the Nuclear Control Institute, 'the MOX option' sends the wrong signal in three ways.

First, this option effectively declares that plutonium has an asset value, and that the energy contained within it should be viewed as a 'national asset' (as the US DOE expressed it) or even 'national treasure' (as the Russians put it), when, in fact, plutonium fuel has been shown to be an economic liability. Second, the MOX option suggests that a commercial plutonium fuel cycle can be effectively safeguarded, when, in fact, it is becoming obvious that large-throughput plutonium plants face daunting safeguard problems. Third, the MOX option would be portrayed as giving credibility to the claim that plutonium recycle in light water reactors (LWRs) is essential to nuclear waste management, at a time when direct disposal of spent fuel is looking increasingly attractive to utilities.

There are other arguments that relate to proliferation using high-level radioactive waste. It is believed that the technologies of Laser Isotope Separation and the Large Volume Plasma Process may permit the mining of weapons materials from any matrix.

There are many international transporting-related issues. It is not uncommon that reprocessing of one country's spent fuel or waste is taken up in a different country. Such movement is often via one or more countries or over the international waters. Regulatory mechanisms, both national and international, have to be in place to guarantee safety of the waste under these conditions.

# Radioactive waste management in India

Just as per capita consumption of electricity is related to the standard of living in a country, the electricity generation by nuclear means can be regarded as a minimum measure of radioactive waste that is generated by a country and hence the related magnitude of radioactive waste management. On the scale of nuclear share of electricity generation, India ranks fourth from the bottom in about 30 countries. As of the year 2000, India's share of nuclear electricity generation in the total electricity generation in the country was 2.65% compared to 75%, 47%, 42.24%, 34.65%, 31.21%, 28.87%, 19.80%, 14.41% and 12.44% of France, Sweden, the Republic of Korea, Japan, Germany, UK, USA, Russia and Canada, respectively. The reactors in operation produce in net Gigawatts (one billion (10<sup>9</sup>) watts) (E) in the latter countries nearly 63, 9,13, 44, 21, 13, 97, 20 and 10, respectively; India's reactors in operation yield 1.9 on this scale (both data are as per IAEA Report of 2000). Hence the magnitude of radioactive waste management in India could be miniscule compared to that in other countries, especially when one takes into account the nuclear arsenal already in stockpile in the nuclear weapons countries. As more power reactors come onstream and as weaponization takes deeper routes the needs of radioactive waste management increase and in this context the experience of other countries would provide useful lessons.

Radioactive waste management has been an integral part of the entire nuclear fuel cycle in India. Low-level radioactive waste and intermediate-level waste arise from operations of reactors and fuel reprocessing facilities. The low-level radioactive waste liquid is retained as sludge after chemical treatment, resulting in decontamination factors ranging from 10 to 1000. Solid radioactive waste is compacted, bailed or incinerated depending upon the nature of the waste. Solar evapora-

	Liquid waste		Solid waste	
Source	Average annual generation (m³)	Specific activity (Bq/ml)	Average annual generation (m³)	Radiation field (mCi/l)
Research reactor	16000	1–3	20–25	0.01-1000
Power reactor				
BWR	26800	50-100	80	0.05-1000
PHWR	26800	0.1-1	100	0.01-1000
Fuel-reprocessing facility	34300	4–20	130	0.01-500
R&D lab	12000	1–4	50	0.01-7000

tion of liquid waste, reverse osmosis and immobilization using cement matrix are adopted depending on the form of waste. Underground engineered trenches in near-surface disposal facilities are utilized for disposal of solid waste; these disposal sites are under continuous surveillance and monitoring. High efficiency particulate air (HEPA) filters are used to minimize air-borne radioactivity. Over the past four decades radioactive waste management facilities have been set up at Trombay, Tarapore, Rawatbhata, Kalpakkam, Narora, Kakrapara, Hyderabad and Jaduguda, along with the growth of nuclear power and fuel-reprocessing plants. Multiplebarrier approach is followed in handling solid waste. Box 5 shows the characteristics of liquid and solid waste generated in India.

After the commissioning of the fast breeder test reactor at Kalpakkam, one is required to reprocess the burnt carbide fuel from this reactor. As the burn-up of this fuel is likely to be of the order of 100 MWD/kg, nearly an order of magnitude more than that of thermal reactors and due to short cooling-time before reprocessing, specific activity to be handled will be greatly enhanced. The use of carbide fuel would result in new forms of chemicals in the reprocessing cycle. These provide new challenges for fast-reactor fuel reprocessing.

#### Concluding remarks

The problems associated with radioactive waste management on a long-term are major ones that humanity has not been able to come to terms with so far. The problem of radioactive waste management has been compared to a Gordian knot. The Gordian knot should not be just sliced through quick and deftly. As American Ambassador Rich III put it, 'The obstacles cannot be over soon or ignored. We must untie the Gordian knot carefully and painstakingly, using all of our resources and democratic institutions wisely and well'.

It is nearly 45 years since the IAEA was founded. Over these years the Agency has deliberated on various issues that confront radioactive waste management and has been providing guidelines and forums for technical and non-technical debates and discussions. As time passes by, new issues crop up, which need to be discussed. One example is how does one 'plan for retirement of nuclear facilities', sometimes referred to as 'decommissioning of facilities'. Similarly changes in concepts of long-term issues on health and safety need to be addressed – 'dose and risk for a remote time in the future are not believable, since habits of human populations are impossible to be predicted'.

All options have not been examined in totality. 'The value of learning by holistic studies of so-called natural analogues is getting appreciated. These are natural systems (such as ore bodies, clay beds and alkaline springs) or archaeological artifacts (Roman glasses, ancient metallic objects and so on) that exhibit some of the key features that repository analysts need to understand. By studying how these systems have evolved over geological time scales, one can gain insights into future repository evolution... The problems will not be solved by throwing unlimited money at them. Some processes take their own time to fructify...'.

- IAEA Bull., 2000, vol. 42, contains a large number of articles related to radioactive waste management. This publication gave an impetus for writing this review.
- Special issue on Radioactive Waste Five articles in Phys. Today, June 1997 and subsequent letters.
- Articles in 'Confronting The Nuclear Legacy' in three parts, Sci. Am., 1996–1998.
- 4. Natarajan, R., in IANCAS Bull., July 1998, p. 27.
- Kumra, M. S. and Bansal, N. A., in Facets of Nuclear Science and Technology, Department of Atomic Energy, Mumbai, 1993.
- 6. Surender Kumar, et al., IAEA-SM-357/38, 1999.
- 7. Many resources on the world-wide-web.

Received 25 May 2001; revised accepted 27 September 2001