Waste Management for Nuclear Power

by P.J.West

The prime objective of waste management is the protection of man and his environment. When waste management is considered in the field of nuclear power, wastes from all sections of the nuclear fuel cycle must be taken into account.

The nuclear fuel cycle encompasses the physical and chemical activities necessary to produce the fuel for use in the reactor; the operation of the reactor for the production of electricity; and the reprocessing of the spent fuel for the recovery of re-usable material. The major steps in this fuel cycle are:

(1) To discover, extract from their place in nature, and purify the naturally-occurring fertile and fissile material;
(2) To convert this material to the proper chemical form for enrichment;
(3) To prepare the required mixtures of fissile and fertile material by either isotopic adjustment or physical blending;
(4) To physically convert the mixture into the required form and containment for reactor operation;
(5) To operate the reactor for the production of electricity; and
(6) To separate and purify the unburned fissile and fertile material in the spent fuel, as well as the fissile material generated in such fuel by reactor operation, for re-use.

Not all reactor concepts require the same fuel cycle. For example, some heavy-water moderated reactors are fuelled with natural uranium requiring no isotopic enrichment, recovery, or recycle of spent fuel. Some water-cooled reactors may use plutonium as well as uranium 235 for the fissile material in reload cores (the second and subsequent fuel cores). Fast reactors use plutonium-239 as well as uranium-235 and uranium-233 as the fissile material. However, since the waste products of the fission reaction are similar for all fissile materials, the waste management problems of the fuel reprocessing step are similar. At the present time the slightly enriched uranium fuelled water-cooled reactor is the system predominantly used throughout the world.

The nuclear fuel cycle is illustrated in **Figure 1.** In the first step uranium bearing ores are removed from the earth in underground or open-pit mines by methods similar to those for extracting many other kinds of metal ores. In general, uranium ores are considered low grade; in the United States the average uranium content of the ores is about one-quarter of one per cent. To minimise the cost of transporting these low grade ores, the uranium mills have generally been built close to the mines. At the mills the ores are crushed and ground, and the uranium extracted with an acid leach. The uranium fraction is generally converted to the oxide form (U_3O_8), and the remainder of the ore is a waste product, generally called mill tailings.

MILL TAILINGS

Since the weight of the uranium removed from the ore is relatively small, the concentration of the radioactivity in the tailings is very much the same as in the ore. About 91,000 tons of tailings arise per year in providing the uranium to fuel 1000 MW(e) (LWR) generating capacity. The concentration of radium-226, the daughter-product of greatest interest because of its combination of long half-life and high specific radiotoxicity, has averaged about 800 picocuries per gram in uranium ores in the United States. In terms of weight this is less than one part in one billion. Because the ores have been ground to the consistency of a fairly fine sand in the milling process, the mill tailings piles are subject to wind erosion. However, since the radioactivity concentration is very low, the effect of such wind erosion is primarily a dust nuisance. The main objective of waste management here is to minimise this dust nuisance. Frequently uranium mills are in regions of very low rainfall, and it is necessary to spray the tailings at active mills with water to hold down the dust. Dikes are used to keep possible water run-off from reaching nearby streams, and to keep occasional high water in the streams from eroding the tailings. In some cases where mills have been shut down, it has been possible to grade the tailings, cover with topsoil, and develop a growth of vegetation which will maintain itself naturally. Under stabilized conditions the radioactive materials in the tailings, with the possible exception of radon, are situated, relative to the environment, similarly to the near-surface ores which have not yet been mined.

Radon is the first daughter-product of radium-226 which, as already noted, is the main radionuclide of interest in the mill tailings. Since radon is a gas it may diffuse outward from radium bearing materials. Radon from typical mill tailings piles, both with or without cover of topsoil, has been found to be undetectable above the general radon background beyond a few hundred metres. However, there is general agreement that because of radon emanations, uranium mill tailings should not be used either in structural materials or in backfill material in connection with buildings intended for human occupancy, and similarly such buildings should not be constructed on top of tailings piles. This leads to a requirement to assure control of such tailings piles for thousands of years.

Continuing in the fuel cycle, the concentrates from the mill are sent to a refinery, where the uranium is extracted with an organic solvent and converted by heating to essentially pure uranium trioxide (UO_3). This material still has its natural isotopic composition of 0.7% uranium-235 and 99.3% uranium-238. Since many reactors are designed to operate with fuel of a higher relative abundance of uranium-235, isotopic enrichment is the next step in the fuel cycle.

ENRICHMENT

The enrichment process in general use is the gaseous diffusion, for which the required feed material is uranium hexafluoride (UF_6), obtained by treatment of the uranium trioxide with hydrogen fluoride and fluorine. The uranium hexafluoride is a solid at room temperature, but can be maintained as a gas by heating, and the gas is forced through a number of porous barriers where the slightly faster diffusion rate of uranium-235 (due to the lower molecular weight) provides a significant enrichment in the uranium-235 content. The residual uranium, which is correspondingly depleted in its uranium-235 content, is presently not a waste but is stored at the diffusion plant for possible later use. The waste management problems at enrichment plants are concerned almost entirely with non-radioactive chemical wastes, and the conservative procedures designed to minimise the loss of uranium from the gaseous diffusion process also serve to minimise the non-radioactive wastes from the operation. The major gaseous and airborne contaminants come from the fossil-fuelled boilers used to produce the process heat. The liquid waste stream contains ammonia, chlorides, fluorides and nitrates generated in process clean-up and auxiliary operations.

The impurities in the liquid waste stream are present in very low concentrations. The non-radioactive wastes from a typical UF_6 conversion plant using either the dry or wet process and from a uranium enrichment complex are given in **Tables 1** and **2** respectively. Radioactive waste production from uranium enrichment and fuel fabrication is difficult to forecast. Whilst the quantities of fuel produced will be a function of the nuclear generating capacity, several other factors will influence the rate of radioactive waste production to such an extent that it is not possible to relate this production to

TABLE 1. UF_6 Conversion Plants – Non-radioactive effluents (in MT/yr) for 5000 MT Uranium/yr.

Constituent	Dry Hydrofluor	Process	Wet	Process
Gaseous and Airborne	Process	Fuel Combustion[1]	Process	Fuel Combustion[1]
SO_x		0.1	18	0.4
NO_x		25.0	60	75.0
Hydrocarbons		5.0		15.1
Fluorine and Fluoride	1.2		1.2	
<u>Liquid</u>				
Sulfate	225		22.5	
Nitrate			6	
Fluoride	481		5	
Cl^-	9		3.3	
Na^+	52		6.7	
NH_3	86		-	
Fe	2		0.5	
K	127		-	
<u>Solid</u>				
Fluoride in CaF_2	250[2]		400	

[1] Natural gas combustion for process heat requirements

[2] CaF_2 bed material

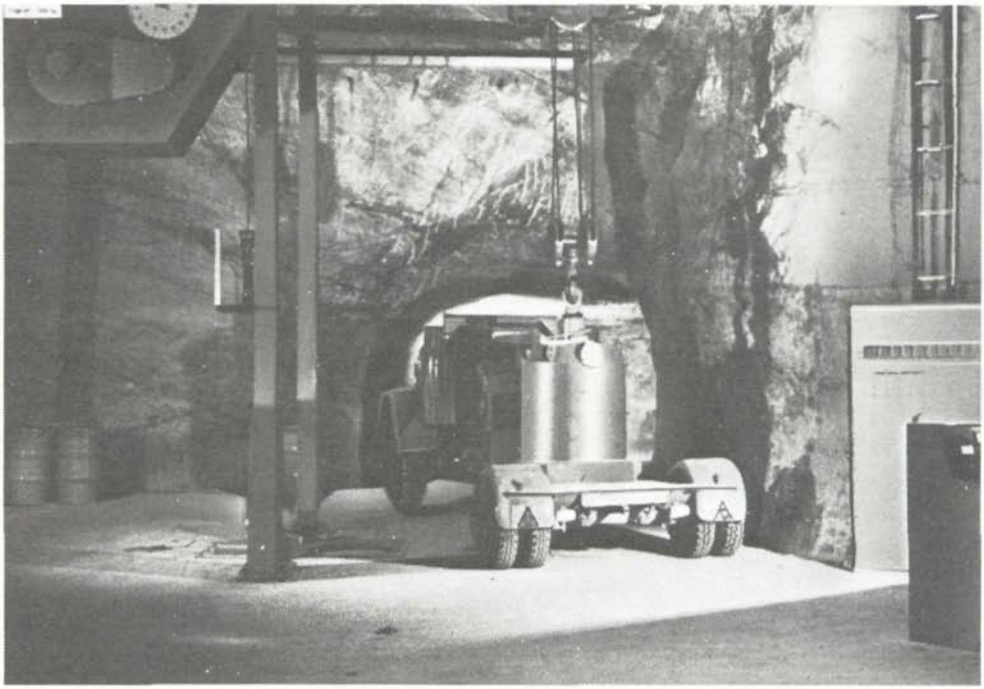
TABLE 2. Isotopic Enrichment Complex – Non-radioactive effluents (in MT/yr) for 10,500 MT SWU/yr

Constituent	Quantity MT/yr	Concentration mg/l
Gaseous and Airborne		
SO_x	740	
NO_x	1390	
Fluorides	45	
Liquid		
Ammonia	16	1.0
Chlorides	150	9.0
Fluorides	16	1.0
Iron	36	2.0
Nitrates	240	15.0

the quantity of fuel fabricated. These factors include improvements in fuel manufacturing technology and also uranium enrichment operations. The probability of plutonium recycle in the fuel for light water reactors will lead to plutonium-239 becoming the main contaminant of concern in the wastes generated by fuel fabrication facilities, due to its high toxicity and exceptionally long half-life of 24 000 years.

The radioactivity induced by neutron capture is the major source of radioactive waste which requires management at the power reactor site. However, far larger quantities of radioactive waste are created in the reactor within the fuel elements themselves, but the problem of management of these wastes arises at the fuel reprocessing plant.

Structural materials used in the reactor and the components which remove heat from its core will corrode and erode only very slightly with time, but enough to create particulates identified broadly as corrosion products. These corrosion products along with other impurities in the coolant circulated through the core of the reactor where they are exposed to neutrons, and neutron capture causes them to become radioactive. The quantities of radioactive materials so formed are small compared with the fission products and consist commonly of radioisotopes of elements such as iron, cobalt, and manganese. Some reactors use boron in the reactor core and core coolant to control the fission process. Neutron absorption by boron leads to the formation of tritium, a radioisotope of hydrogen. Tritium formation similarly can result where water is used as a coolant, through conversion of deuterium, a natural isotope of hydrogen present in the water. In gas cooled reactors, cooled with carbon dioxide, the activation products include argon-41 and carbon-14.



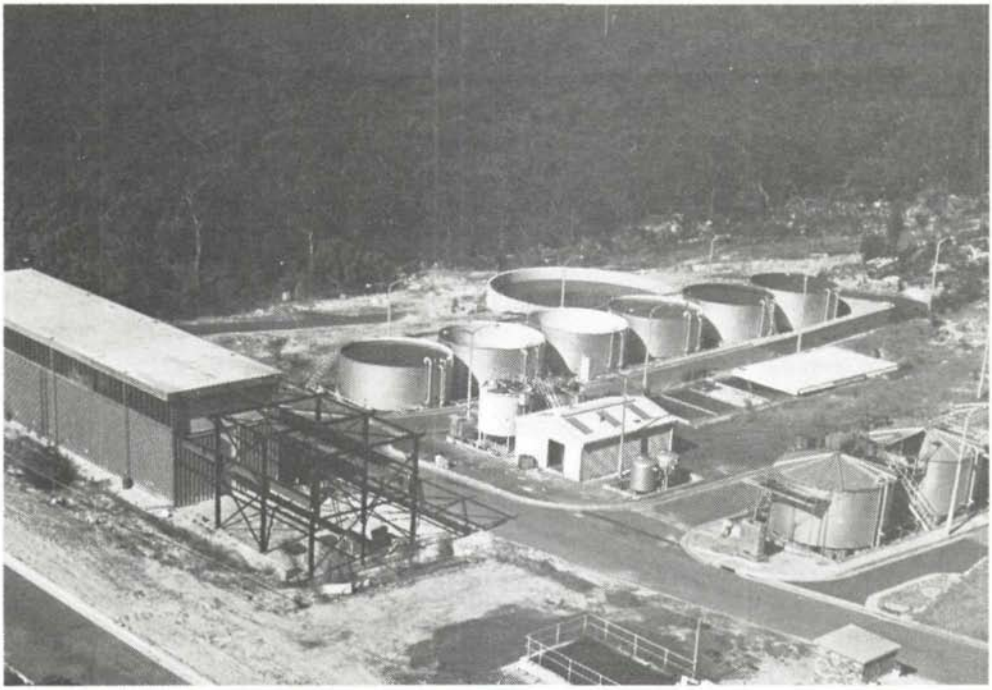
Trucks leave the former north German salt mine, Asse 11, which the Federal Republic is using as an underground storage site for its low and medium level radioactive wastes.

EFFLUENTS

Although the kinds of radioactive wastes produced as by-products of the fission process are basically the same for all uranium fuelled reactors, the characteristics of effluents from power plants can vary appreciably, depending on the reactor coolant and steam cycles used. The radioisotopes in the effluent streams in turn influence the design of waste treatment systems. The objective in plant design is to process and recycle waste streams so as to minimise both the volume and radioactivity of effluent wherever practical. Releases to the environment are controlled both by batch processing of effluents and by monitoring before discharge, to ensure that no release exceeds the established permissible limits.

Liquid waste management systems employ four basic treatment techniques to reduce levels of radioactivity. These are delay and decay; filtration; evaporation; and ion exchange.

Delay and decay refers to the storage of waste for long enough time to permit some of the radioactivity to decay. This technique has its main value in respect of radioisotopes of short half-life. The waste management techniques in use for gaseous wastes are delay and decay; filtration; and low temperature absorption on charcoal. The usefulness of delay and decay as a means of reducing activity levels in gaseous wastes depends on the particular isotopes present. Gases are then filtered and released through stacks to the atmosphere. Filters collect radioactive solid particles formed when a gaseous parent nuclide decays to a particulate radioactive daughter or becomes attached to particles; and when particles of dust are carried by the air stream through the reactor core. Specially treated charcoal filter beds may be used to remove iodine. Solid waste arisings



The low level radioactive waste treatment plant installed at the Lucas Heights nuclear research establishment of the Australian Atomic Energy Commission. Photo: AAEC

may include used nuclear reactor equipment which contains induced radioactivity by neutron capture, used high-efficiency particulate filters from ventilation exhaust systems, precipitates, sludges, or ion exchange resins containing radioactive materials arising from the treatment of liquid effluents, and used protective clothing or other materials required for occupational safety or contamination control reasons. In general the radioactive waste management problems associated with reactor operation are relatively minor compared with those that are associated with the reprocessing of the nuclear fuel. There is, however, another waste arising from nuclear reactor operation which is of concern in relation to its possible environmental effects, and that is the dispersal of the waste heat.

WASTE HEAT

No method of converting heat to electricity uses all the heat which is available. Modern steam turbines operating with fossil fuels and using high pressure steam at high temperatures attain thermal efficiencies of 40% or more, and in these plants part of the excess heat is released to the atmosphere in the flue gases. In nuclear power plants essentially all of the excess heat is transferred to the cooling water. As a result, a nuclear power plant in which the cooling water is only passed through the cooling circuits once, will discharge about 50% more waste heat to the receiving waters than a fossil fuelled power plant producing the same amount of electricity. Gas-cooled and liquid-metal-cooled advanced reactors of the future are expected to attain higher thermal efficiencies.

There is no doubt that the discharge of waste heat into public waters can modify the aquatic environment. Apart from considerations of temperature effects in the receiving water, the dangers to the food chain by damage to biota through mechanical and thermal shock in the passage of the cooling water through the condenser system, may also provide a disturbance to the aquatic ecology. Knowledge of the aquatic life present in the receiving waters, coupled with the use of engineering techniques designed to minimise the impact of the release of waste heat, can enable power plants to meet the desired standards of water quality. If there is not enough water available to meet these standards with a once-through cooling system, then provision must be made for alternative systems such as cooling towers and cooling ponds to recycle the cooling water and thus reduce the effects of waste heat to acceptable levels. Nevertheless large cooling towers also have their environmental impacts, which in addition to their aesthetic aspects may include ground level fog, clouds and precipitation, shadowing, and synergistic effects through the plumes mixing with industrial stack effluents containing oxides of sulphur and nitrogen.

FUEL REPROCESSING

The last sector of the nuclear fuel cycle, the reprocessing of the irradiated nuclear fuel, is associated with the management of more than 99% of all the waste radioactivity arising from nuclear power generation.

Fission products build up in the fuel elements within the reactor, eventually absorbing neutrons to such an extent that the fission process is interfered with. The fuel elements are therefore removed from the reactor well before all the usable fuel has been burned, and are sent to reprocessing plants. The main objective of reprocessing is the safe and efficient recovery of plutonium (which is produced in the reactor) and unburned uranium in sufficient purity to re-use in the fuel cycle.

In considering the waste management problems associated with fuel reprocessing, it will be helpful to summarize the main types of operation, so that the origins of the various wastes are made apparent:

(1) Disassembly operations are sometimes necessary to remove external parts of the irradiated fuel assembly by remote handling methods prior to starting the reprocessing operation. This gives rise to an accumulation of metallic waste, which, as a result of neutron activation in the reactor, is usually radioactive.

(2) Decladding of some fuels, especially uranium metal fuels, is carried out either by chemical dissolution of the cladding material or by a mechanical process. The dissolution process gives rise to a liquid effluent, whereas the mechanical process leads to a solid waste in the form of metal swarf. In each case the cladding activity, caused mainly by diffusion of fission products from the fuel into the cladding and by neutron activation, gives rise to a radioactive waste.

(3) Other fuels, especially those made from uranium oxide, are fed to a shear and chopped into small sections which drop into a dissolver vessel. This operation may release small quantities of gaseous effluent, including radioactive noble gas fission products.

(4) Metal fuels after decladding and oxide fuels after shearing are dissolved, usually in boiling aqueous acid. This gives rise to a gaseous effluent containing radioactive

fission products such as the noble gases krypton and xenon, together with iodine. Sometimes the fuel and cladding are dissolved entirely, but more often only the irradiated uranium oxide is dissolved, leaving a residue of leached metallic hulls to be disposed of as radioactive waste.

(5) Chemical processing is carried out in a series of solvent extraction and sometimes ion exchange treatments with intermediate chemical conditioning stages. At the outset the bulk of the fission products are separated from the uranium and plutonium to give an intensely radioactive waste solution, which must be stored in some form or other after treatment and concentration. As the processing for recovery of the two major end products, uranium and plutonium, proceeds, a number of liquid effluents arise containing the remaining traces of fission products and trace amounts of uranium and plutonium. Some solid wastes, such as ion exchange resins, used filters, etc. of moderate activity level, also arise from these operations.

Having summarized briefly the origins and types of radioactive waste arising from fuel reprocessing, the non-radioactive wastes are also worthy of mention in view of their potential environmental impacts. The typical non-radioactive wastes arising from a reprocessing facility based on 900 MT/year throughput of uranium are given in **Table 3.**

TABLE 3. Fuel Reprocessing Plant – Non-radioactive Effluents (MT/yr)

Constituent	MT/yr
<u>Gaseous and Airborne</u>	
SO_X	160
NO_X	185
Hydrocarbons	0.5
Fluoride	28
<u>Liquid</u>	
Na^+	137
Cl^-	6.3
$SO_4^=$	11.3
NO_3	21.6

SOLIDS, LIQUIDS AND GASES

The management and control of the radioactive waste are discussed under the three collective headings, solids, liquids and gases.

The principal solid wastes are the metallic discards, cladding swarf and leached sections of fuel can. Usually these wastes are too radioactive for disposal by simple ground burial, although this procedure has been adopted at some facilities where it has been established that suitably impervious geologic strata exist. More often these metallic wastes are stored in concrete silos on the site, designed and operated in such a way as not to preclude the possibility of ultimate removal and disposal after a very long decay

period. Other solid wastes are monitored and segregated into convenient types, such as combustible and non-combustible, low-level and medium level activity. A variety of disposal techniques have been adopted for these wastes in different countries.
For the lower levels of activity burial in large trenches on the site has been used; long-lived activity is limited in these disposals so that ultimate release of the site is feasible. Regular monitoring of water draining from such sites is carried out to demonstrate continued safety of the procedure. Another technique for the disposal of low-level solid waste, which has been used by some countries, is sea disposal under carefully controlled conditions; the waste is placed in drums which are encased in concrete and dumped in selected deep ocean areas at a depth of at least 2700 metres. Such dumping has often been carried out as a combined international operation. Combustible waste has been incinerated at some sites, with special attention being paid to the cleansing of the gaseous combustion products by conventional scrubbing and filtering techniques. The resultant ash can either be subjected to chemical recovery treatment or stored economically because of its small bulk. Medium-level solid wastes are usually stored on a long-term basis to allow their activity to decay.

The principal gaseous wastes in reprocessing arise from the shearing of oxide fuel, from the fuel dissolving operation, and from the air used to ventilate facilities such as high activity liquid storage tanks. Particulate and entrained activity may be involved as well as iodine-131, iodine-129 and radioactive noble gases such as krypton-85. Invariably fuel is stored sufficiently long before reprocessing to allow most of the short-lived iodine-131 (half-life 8 days) to decay so that this radionuclide rarely presents a significant problem, but specially designed absorption filters are used to trap radioiodine. Particulate and entrained activity is kept to a low level by using specially designed gas cleaning equipment including scrubbers and filters. Release to the environment is ultimately through high stacks fitted with monitoring equipment which registers activity levels and flow rates so that activity levels in discharged gases may be known and recorded. As a final check at most reprocessing sites a programme of environmental monitoring is carried out in the area surrounding the plant, often extending for a number of kilometers. The samples taken and examined include those indicative of potential public exposure, including radioiodine and radiostrontium in milk.

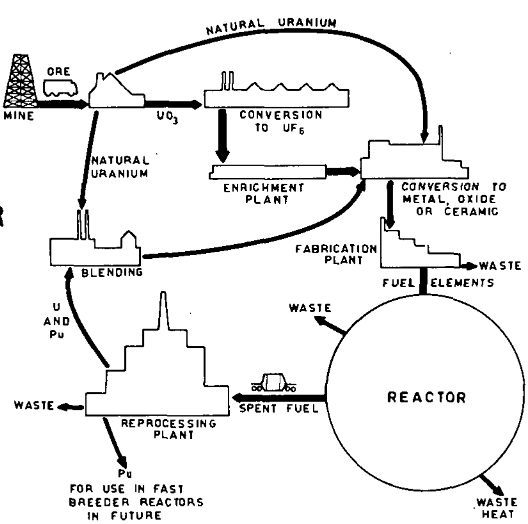
There are three major categories of liquid wastes from reprocessing: low-level wastes such as fuel storage pond water, condensates from evaporators and many plant effluents; medium activity wastes such as those arising from chemical decanning operations and some plant effluents; and high activity wastes containing the bulk of the fission products (usually more than 99.9%).

Large reprocessing plants produce several cubic meters per day of low level aqueous wastes. After some treatment which varies from simple storage to permit radioactive decay to sophisticated chemical processing, such wastes are usually released to rivers or the sea under carefully controlled conditions in accordance with regulatory standards. Medium activity wastes are very variable in composition, but usually contain significant quantities of dissolved salts. They are treated by a selection or combination of methods such as evaporation (with transfer of the concentrate to high activity waste storage), decay storage and precipitation (with the settled sludge being retained as solid waste). The major volume of such waste is thus transformed into low level liquid waste.

The high activity waste has the bulk of the fission products generated in power reactors contained in aqueous solutions of nitrate salts of various metals after reprocessing. The composition of the waste varies depending on the reagents used in the processing, and whether the fuel has been leached from the fuel cladding or whether cladding and fuel have been completely dissolved. Invariably the first stage of treatment is concentration by evaporation, but the degree of concentration varies considerably depending on the process flowsheet used. If only small amounts of extra salts have been added and the cladding has not been dissolved, then a volume reduction to about 50 litres per tonne of uranium fuel is achieved, but in a number of plants 450 - 900 litres per tonne of fuel result. This intensely active liquor is stored in thick walled steel tanks contained in concrete cells up to 1.5 m thick, themselves lined with steel. In order to remove the heat of the decay of the fission products, cooling coils are incorporated in the tanks, and sometimes a means of keeping precipitated solids in suspension, such as air jets, is included. Sufficient spare tank capacity is obtained to accept the contents of a leaking tank. Whilst it is considered safe to store these concentrates as liquids in such tanks for many years, it is almost universally accepted that some form of solidification is preferred for storage. As already mentioned, different processes give rise to rather different high activity liquid wastes, and certain solidification techniques that have been developed are more suited to some wastes than to others.

Generally, all solidification processes involve heating the wastes to temperatures between 400°C and 1200°C, which drives off all the volatile constituents, mainly water and nitrates, leaving a calcined solid or a melt that cools to a solid. In most cases melt-making additives are included with the heated waste so that glass-like products result. Ideally the solidified wastes should have good thermal conductivity, low leachability, high chemical stability and radiation resistance, and mechanical strength. After solidification the wastes must be suitably contained. Interim storage of the solid on the site of the fuel reprocessing and waste solidification plant will probably be necessary, to allow

THE NUCLEAR FUEL CYCLE



radioactive decay of most of the activity of radionuclides with short and intermediate half-lives. In order to dispose of the heat from radioactive decay, the storage facilities will need to be designed to provide cooling, using either air or water. In some countries final disposal of the solidified waste in deep geologic formations such as salt domes is now being examined. The technology currently available for reprocessing of irradiated fuel

TABLE 4. Quantities of Solid Wastes Containing Fission Products Estimated Annual Rate

Solid Waste	1000 MW(e) LWR	1000 MW(e) LMFBR
Produced at Reactor Site		
Low-Level		
Cubic metre/yr	56 - 110	28 - 56
Kilograms/yr	60,000 - 100,000	30,000 - 50,000
Number 240 litre drums/yr	270 - 540	135 - 270
Storage site area used — square metre/yr	165 - 330	83 - 165
Produced at Reprocessing Plant		
High-Level Vitrified		
Cubic metre/yr	2.5	2.0
Kilograms/yr	5000	4000
Canisters/yr	60	40
Repository space required — square metre/yr	1100	750
Cladding Hulls		
Cubic metre/yr	1.7	5.5
Kilograms/yr	7600	30,000
Number of 150 litre drums/yr	15	50
Storage area/yr	a	a
Low-Level Solids		
Cubic metre/yr	17 - 115	56 - 340
Kilograms/yr	30,000 - 65,000	100,000 - 200,000
Number 250 litre drums/yr	80 - 540	270 - 1600
Storage site area used — square metre/yr	93 - 460	185 - 930
Produced at Fuel Fabrication Plant		
Pu Contaminated Wastes		
Cubic metre/yr	340[b]	230
Number 250 litre drums/yr	1500[b]	1000
Repository volume — cubic metre/yr	600[b]	400

[a] Depends on whether storage is required at commercial site or Federal Repository.
[b] Assumes Pu recycle.

does not permit the quantitative separation of the actinides from the fission product wastes, so that these wastes do contain some long-lived radionuclides such as plutonium-239 (half-life 24 000 years). Therefore, solidified wastes currently being obtained from the high-level waste concentrates will need to be isolated from the human environment for many thousands of years.

The quantities of radioactive waste arising per 1000 MW(e) generating capacity per annum for LWRs, which will be the predominant reactor type at least up to 1990, are shown in **Table 4.** Since the liquid metal fast breeder reactor is likely to be the reactor type to take over the major role after 1990, comparative figures are also given in this table of the wastes arising per 1000 MW(e)/year for the LMFBR.

The problems of managing these high-level wastes are recognised and new developments are being carefully evaluated. The ultimate techniques to be used will be selected only after there is assurance that a proper balance has been achieved between a sufficiently high probability of success and the risks that can be identified.

The total risk associated with the management of radioactive wastes is the sum of the risks encountered in association with each step. The main goal is to reduce the cumulative risk to the lowest practicable level. As a result, the steps employed in waste management programmes may differ from country to country, depending on the magnitude of the risks associated with each process used.

Wylfa Nuclear Power Station in Anglesey, North Wales, which won a commendation in the 1973 Financial Times newspaper award for an outstanding work of industrial architecture. The power station was one of six finalists among the applications. In their report on Wylfa nuclear plant, the jury commented: "This project demonstrates an unusually sensitive approach in a coastal landscape of great beauty. A massive nuclear power station, through the restraint and care of the architects and landscape planners, has turned potential disaster into a near advantage. These huge buildings have a unity and simplicity that somehow relates them to the quality of the landscape, aided by the muted use of colour and imaginative form."