Radioactive Waste Management

by William L. Lennemann

The radioactive elements (radionuclides) cannot be destroyed by any known chemical or mechanical process. Their ultimate destruction is through radio-decay to stable isotopes or by nuclear transmutation by bombardment with atomic particles. Consequently, radioactive waste management consists of controlling and reducing the radioactive releases to tolerable levels, removing from effluents and wastes those radionuclides of concern, concentrating them into a form which can be stored or disposed of in such a manner that they do not later appear in hazardous concentrations in the biosphere.

It would take volumes to discuss the various aspects of radioactive waste management and cover the technologies involved. Volumes have been written. Consequently, it would be presumptuous to expect more than a brief mention of what is occurring in the radioactive waste management area in a review article. Undoubtedly, many aspects will be overlooked or missed in this status report. Hopefully, it still will show that the scientists, engineers and operators involved with the nuclear fuel cycle are, and will be, meeting the technological requirements for the safe management of wastes contaminated with radionuclides as the need arises.

BACKGROUND

Radioactive waste management involves two fundamental approaches: the radioactive materials can be either released or discharged to the environment, or they must be confined and isolated from the biosphere until the noxious radionuclides have decayed to innocuous concentrations.

Releases of radioactivity to the environment generally occur as liquid or gaseous discharges (effluents) from nuclear facilities. The amount of radioactivity which can be released is based on allowable exposures to population groups and is controlled by national regulations and guidelines, usually based on recommendations of the International Committee on Radiation Protection.

There are very few instances where radioactive effluents from nuclear facilities can be released without some form of control or treatment to remove excessive radioactivity. Most of the radioactive wastes arising from the operation of nuclear fuel cycle facilities require processing to concentrate the radioactive elements into a smaller volume which can be more conveniently handled, thereby permitting the release or disposal of the bulk of the processed material.

Radioactive waste management involves the use of normal industrial operations and techniques adapted to cope with the barriers needed to protect the workers and operations from excessive radiation and contact with the radionuclides. Consequently, very often the

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equipment and its operating procedures become rather sophisticated to permit operation and maintenance behind the protective barriers. Actually, most research and development activities involving technology for the management of radioactive wastes are concerned with adapting normal industrial operations or laboratory techniques to the handling of radioactive materials.

It is the consensus amongst those involved with, or knowledgeable about, radioactive waste management, that technology and flow schemes have now been developed to the extent that they are workable. The basic knowledge required to collect, treat, package and store safely all nuclear fuel cycle wastes of concern is available. Furthermore, in many cases, optional technology exists for different situations or economic optimization.

Consequently, from a technical standpoint, it seems that lack of appropriate methods and technology for handling the radioactive waste need not become a bottleneck for the implementation of nuclear power. On the other hand, some of the required radioactive waste management technology still remains in its developmental stage. Much remains to be done in working out the engineering and design details, satisfying appropriate regulatory requirements, and adapting the technology to actual operating conditions and controls. These last remarks apply principally to those radioactive wastes arising from the reprocessing of irradiated fuel which essentially contain all the radioactive products of nuclear fission.

MILL TAILINGS

Uranium mill tailings do not present a serious radiation hazard in open, well-ventilated areas. The principal concern involves the possiblity of hazardous concentrations of radon gas from the decay of the relatively small amounts of radium-226 in the tailings.

Tailings disposal areas can be appropriately located, managed and isolated. The technology exists to stabilize these tailings with a ground cover and vegetation and, if necessary, to protect them in other ways, in order to prevent their disperson by wind, water and man. Furthermore, such tailings areas can (and should be) recorded with the local and state land authorities, with restrictions in perpetuity being placed on their use.

EFFLUENTS FROM URANIUM REFINING AND ENRICHMENT

In uranium refining and enrichment operations, the toxic, non-radioactive chemicals in the liquid and gaseous effluents are more of a problem than the radioactive contaminants. There are proven techniques and industrial practices available for preventing environmental pollution with the organic solvents, nitrates, sulfates, ammonia, chlorides, fluorides, fluorine and other chemicals used in the refining and enrichment processes. The depleted uranium from enrichment plants is currently not considered a waste and is recovered and stored.

However, the front end equipment of both uranium refineries and the processes for converting the refined uranium to uranium hexafluoride do eventually become radioactive from a build-up of the traces of radioactive elements still present in the concentrates and refined uranium. These radionuclides adsorp or plate on equipment surfaces and especially concentrate in the "ash" remaining from the uranium hexafluoride process. Such radioactive equipment and material generally are disposed of by suitably controlled land burial. Ocean dumping also has been used.

URANIUM FUEL FABRICATION

The main waste management concern in the fabrication of uranium fuels comes from the non-radioactive elements and chemicals used in their manufacture. Uranium in waste is recovered and concentrations of uranium in effluents form uranium fuel fabrication plants are minimal. Normally, industrial non-radioactive waste management techniques and safety practices are used in uranium fuel fabrication and, generally, this is all that is needed. Uranium contaminated equipment can be readily decontaminated for safe disposal or possible reuse.

WASTES WITHIN THE NUCLEAR FUEL CYCLE

The radioactivity induced by neutron capture is the major source of radioactive waste that requires management at the power reactor site. This neutron capture occurs in the corrosion products and other impurities in the coolant circulating through the reactor core, and in the structural components of the reactor that are exposed to high radiation levels.
In addition, small quantities of radioactive fission products occur in the reactor coolant and fuel storage basin water as a result of an occasional fuel cladding failure.

The radioactive contaminants of main concern are fission products resulting from nuclear fission in the fuel elements. These fission products are released at the fuel reprocessing plant, where the fuel cladding is either chemically or mechanically breached and the fuel is dissolved. The soluble fission products dissolve into an aqueous solution, along with the plutonium and unburned uranium. Gaseous fission products also are released to the plant off-gas systems during the breaching and dissolving operations.

The radionuclides remaining in the solution after recovery of the uranium and plutonium are the fission products and the actinides, also called the transuranics, which include the unrecovered plutonium and the heavier elements (Np, Am, Cm, etc) of the actinide series formed by neutron capture. Since these transuranics decay through alpha emission, the collective term alpha-emitters is commonly used. Waste contaminated with the alpha-emitters is called alpha-contaminated or alpha-bearing waste.

The fission products and alpha-emitters contaminate all materials with which they come in contact and this contamination is passed along the contact chain. Hence, the formation of radioactive waste from what otherwise would be normal industrial waste. The extent to which such contaminated materials can be suitably decontaminated for some purpose only results in additional radioactive waste arising from the decontamination process.
The materials handling and fabrication involving liquid and solid forms of fission products and transuranics, especially for plutonium in the fabrication of plutonium and uranium-plutonium oxide fuels, results in contaminated equipment and ventilation systems, all to be dealt with.

GASEOUS WASTES

The gaseous effluents from reactor operations may contain radioisotopes of the noble gases (principally, argon-41, krypton-85 and xenon-133), the radioiodines-129 and -131, tritium and oxides of carbon-14. However, as mentioned, the main release of gaseous radionuclides occurs when the spent nuclear fuel is breached and dissolved at the fuel reprocessing plant. There, the major releases are krypton-85, xenon-133, the radioiodines-129 and-131 and tritium. Xenon-133 with its relatively short (5.3 day) half-life is not anticipated to be a problem, with methods available for delay and decay, if necessary, prior to its release. The radioiodines can be eliminated from gaseous waste streams by counter-current scrubbing of the gas with aqueous solutions of caustic, mercuric nitrate, nitric acid, or by chemical adsorption on zeolites treated with silver or on other metals with an affinity for iodine. Another method is by adsorption on charcoal, which also can be impregnated with chemicals that will take up the iodine complexes.

While the extent to which tritium, krypton-85 and carbon-14 can be released into the atmosphere without becoming a matter of global concern has not been definitely established, analyses have indicated that this should not be a problem before the end of the century. Nevertheless, cryogenic (low temperature) separation methods are becoming available for removal of krypton-85 and xenon-133, if necessary. Other removal techniques, such as absorption in liquid fluorocarbons, adsorption on charcoal or other solids, and diffusion through membranes are under development.

So far, no practical process has been found for the removal of tritium from waste effluents since much of the tritium that is released is highly diluted as tritiated water. A promising approach for tritium removal in fuel reprocessing is to confine, oxidize and collect the gaseous effluents from the shearing and other reprocessing plant head-end operations, converting the tritium (T_2) to T_2O (probably THO) at a tritium concentration of 1 to 5%. Other approaches being investigated are the adsorption of tritium on vanadium oxide, and isotopic separation processes such as high pressure and temperature diffusion through palladium films and the use of molecular sieves. Selective solvent extraction techniques also are being considered.

Carbon-14 can be removed in the oxide form from gaseous effluents by caustic scrubbing or adsorption on solid alkaline media, if the need arises. Radioactive particulate matter and aerosols in gaseous effluents can be removed by high efficiency filtration techniques.

LIQUID WASTES

Radioactive liquid wastes are loosely classified as high-level, intermediate-or medium-level and low-level, based primarily on their radioactivity levels. High-level waste has been generally accepted as the raffinate (liquid effluent) from the first cycle of fuel reprocessing operations. The raffinate contains over 99.9% of the non-gaseous fission products, the unrecovered plutonium and the higher actinides (transuranics). The distinction between medium- and low-level wastes is rather arbitrary and, in addition to their radionuclide content, the classification generally depends upon the extent to which they must be isolated and/or processed as compared with being released to the local surroundings. While medium- and low-level liquid wastes vary considerably with respect to their transuranic and fission product content, nevertheless there is a distinct difference between their treatment processes and those for high-level waste. Consequently they can be considered on the basis of this distinction.

As mentioned, high-level liquid waste is essentially the fuel reprocessing, first cycle raffinates. High-level liquid waste also can include concentrates of fission products and transuranics resulting from the processing of medium- and low-level liquid wastes.

High-level liquid waste is stored in suitably designed, cooled, stainless steel tanks, generally in steel-lined vaults. This method is considered safe for long periods although it requires close surveillance. However, it is the general opinion that these wastes should be converted to a solid form for long-term isolation from the terrestrial biosphere.

Several approaches have been developed for the solidification of high-level waste, including the use of fluidized beds, stirred beds, rotary kilns, sprays and pots. They all essentially involve heating the waste to between 400° and 1200°C, driving off all volatile constituents and leaving a calcined solid. However, most calcined wastes are relatively soluble and usually not considered a suitable product for disposal. Consequently, in most cases, borosilicate or phosphate glass forming constituents either are put into the calcining stage or are added as a second stage of the solidification process, incorporating the calcined waste into a glass melt, which cools to a vitrified product with leaching rates similar to those of Pyrex glass. There are at least 15 approaches to producing essentially the same vitrified product. However, with the exception of the French AVM process, which is scheduled to start full scale industrial operations at Marcoule in 1977, all the other processes have not been tested beyond the developmental stage. A novel approach under development at Eurochemic incorporates the radioactive calcine or glass into metal matrices.

Intermediate-level liquid wastes generally are characterized by smaller volumes and higher concentrations of radionuclides as compared to low-level liquid wastes. However, as indicated, there is no universally accepted distinction between the two. Intermediate-level liquid wastes can arise from spills, off-gas scrubber solutions, ion-exchange regenerates, cask and plant decontamination solutions, plant solvent washes, chemical decladding solutions, the reffinates from uranium and plutonium purification cycles and some laboratory wastes. Examples of low-level liquid wastes are evaporator and concentrator condensates, laundry wastes, condensates from process vessel ventilation systems and possible radionuclide leakage into cooling water or process steam.

All water soluble radioactivity released during reactor operations is introduced originally into either the primary coolant or the fuel storage basin water, mostly from fuel cladding failures. Radioactive particulate products of corrosion also are present. This radioactivity then spreads to other parts of the reactor system by leakage or treatment of these streams; for example, ion exchange regenerates and evaporator slurries. Intermediate- and low-level liquid waste streams arising from fuel reprocessing have been enumerated in the preceding paragraph. Besides decontamination, scrubbing and washing wastes, a mixed oxide ($P_uO_2 + UO_2$) fuel fabrication plant also generally has liquid waste streams from processing scrap material for plutonium recovery. These streams can be considered either intermediate- or low-level material, depending on their characteristic actinide content.

Basic treatment techniques available to reduce levels of radioactivity in liquid waste streams are filtration and centrifugation (to eliminate radioactive particulates), evaporation, ion exchange, flocculation, and precipitation. Seepage ponds also can be used for liquid wastes containing very low concentrations of the shorter-lived fission products. Membrane concentration techniques are being examined but not yet generally used. The products of the foregoing basic treatment techniques are liquid concentrates and sludges which are "conditioned" to a solid form for either storage or disposal. Incineration and steam distillation techniques are available for the treatment of radioactive organic oils and solvents. Also, it is possible to reduce them to a solid state by the use of certain oil-absorbing solid materials.

SOLID WASTES

Besides the solid equipment and materials contaminated with either induced or contact radioactivity, solid radioactive wastes also include the solidified high-level liquid waste, "conditioned" concentrates from intermediate- and low-level liquid waste, loaded or spent ion exchange resins, and gaseous radionuclides incorporated into some solid medium for storage or disposal. Solid wastes fall into one of two categories, combustible or noncombustible. Depending on the facility, combustible and noncombustible waste may be mixed, and may require sorting prior to treatment and/or packaging for storage or disposal.

Combustible solid radioactive waste consists of a large variety of materials such as paper, rags, absorbent cotton, plastic sheeting, protective clothing gloves, rubber shoes, wood, cardboard, organic ion exchange resins, filter aids, combustible high efficiency filter media, etc. Since much of these waste materials is collected as general trash, it generally is sorted prior to incineration. Glove box operations for plutonium purification and fabrication introduce rubber or plastic materials.

Dry, conventional incineration techniques, although successfully practised, still involve problems of total combustion and off-gas cleaning. Other incineration techniques under development involve agitated hearths, cyclone furnaces and fluidized beds. Wet oxidation techniques involving acid digestion, now under development, and pyrolytic processes appear to be promising. Also the incorporation and incineration of combustible waste in molten salt mixtures is being examined.

While failed equipment may offer more difficult problems became of their large size and high level of radiation, there also is the steady stream of miscellaneous noncombustible radioactive waste from each type of nuclear fuel cycle facility. While the primary constituent of noncombustible solid waste is metal, including fuel cladding, other materials such as glass and concrete are significant. Combustible materials, such as organic ion exchange resins, plastics, grease and floor sweepings are often included with noncombustible waste.

Typical large items of equipment from a fuel reprocessing plant may consist of dissolvers, solvent extraction columns or units and concentrators which might be up to 3 metres in diameter and 10 metres in height. Noncombustible waste from reactors can include large components from the reactor core. Many of the large equipment items must be dismantled, sometimes prior to removal from the system. The normal approach is to flush and decontaminate such equipment to the extent possible. Both fuel reprocessing plant and reactor equipment may be highly radioactive, requiring remote handling and personnel shielding equivalent to several feet of concrete. Such equipment may be stored until the radioactivity has decayed to lower levels.

Failed equipment from plutonium processing and mixed oxide fuel fabrication operations, such as glove boxes, may have low enough radiation levels to permit dismantling without shielding or with light shielding for the workers. However, the presence of plutonium requires elaborate precautions to prevent its spread to the environs.

Generally, noncombustible waste is packaged with appropriate shielding. Where possible, the waste can be crushed or melted for volume reduction. Smaller units of noncombustible wastes, such as incineration ash, ion-exchange resins, and sludges and residues from the treatment of intermediate- and low-level liquid wastes are incorporated in concrete and bitumen. Ureaformaldehyde resins, and salt matrices and other absorbants and packing materials also are being used.

STORAGE

Storage implies easy retrievability. Storage facilities for radioactive wastes must prevent radiation exposure to man and provide physical confinement. Continuing surveillance is required to assure that these two conditions are being met.

As previously indicated, it has not been necessary to store radioactive gaseous materials nor is it likely to be for some time to come. However, they can be compressed and stored in metal cylinders. Techniques are being developed to incorporate them within solid matrices and adsorbants as a protection against their sudden release should a container fail. Radioactive liquid wastes can be conveniently stored in tanks or other containers. Storage methods for radioactive solid wastes ranges from the use of concrete or earthen vaults for highly radioactive materials to simple fire-proof structures for low-levels of radiation. Underground mines and tunnels in dry geologic formations also are being utilized. The experience gained from storing irradiated fuel elements is applicable to the storage of solid high-level waste in water basins or air-cooled vaults.

DISPOSAL

Disposal implies a final resting place or repository with no intent of retrieving the material. Furthermore, once the safety and confinement of the disposal method has been assured, surveillance of the site can be minimal or even discontinued. It is generally accepted that, for disposal, it is better for all radioactive waste to be in a relatively insoluble solid form which is less susceptible to dispersion from a disposal site than a liquid or a gaseous form. Also, the method of packaging can be used as a barrier between the contents and its surroundings. On the other hand, the USSR has demonstrated the safe injection of intermediate- and low-level liquid wastes into porous formations in deep geologic structures. In the USA, Oak Ridge National Laboratory is demonstrating the injection of a mixture of liquid waste and solidifying agents into shale seams by hydrofracture.

Regardless of certain attractions for using outer space, disposal of radioactive solid wastes will be terrestrial for some time to come. Land burial and sea dumping are being utilized for the disposal of intermediate- and low-level solid wastes. Land burial techniques range from the use of concrete trenches and vaults, which may then be filled with concrete, to direct burial of the packaged waste in trenches, pits or landfill. Protection against migration of the radionuclides into ground-water is of paramount concern. Sea dumping requires suitable packaging and preparation to withstand or equalize the water pressure on the container and sufficient weight to allow it to sink to the ocean depths and remain there. The amount of radionuclides disposed of at any one site (land or sea) is carefully controlled so as to not exceed the environmental capacity. The extent to which solid waste contaminated with the longer-lived alpha-emitters (alpha-bearing waste) can be disposed of at land burial or sea dumping sites still is under study which includes their concentrations in the waste as well as total quantities.

The disposal of suitably packaged low- and intermediate-level solid wastes in an underground salt formation is being demonstrated in the Federal Republic of Germany. Many countries now are considering a similar approach.

The activation product of most concern found in solid waste is cobalt-60 with a half-life of 5.3 years. Consequently, highly irradiated reactor components generally will have decayed to innocuous levels within 150—200 years. For this relatively short time period, land burial and properly packaged sea disposal would seem to be suitable.

On the other hand, wastes contaminated with the longer-lived fission products may require from 600 to 1000 years before they do not constitue a radiological hazard. Wastes contaminated with plutonium-239 (a half-life of 24,300 years) can require isolation periods of up to 500, 000 years. Other transuranic actinides can require more or less such geologic time periods before their concentrations can be considered tolerable.

Currently, there has been no method demonstrated for the disposal of the solidified high-level and other wastes significantly contaminated with the alpha-emitters, possibly because there has been no need for such facilities. On the other hand, public and political concerns regarding this question can no longer be ignored. Many countries now are examining the possibilities of disposing of the solidified high-level and alpha-bearing wastes into suitable underground geologic structures (geologic disposal) which is the only option available with today's technology. Salt, clay and crystalline rock formations are being considered. One can expect that within the next five to seven years, several demonstration projects for geologic disposal will be operating. Disposal into the deep ocean sediments or into geologically stable areas of the sea bed has been proposed, using the technology developed recently by exploring and developing off-shore oil deposits. However, it will be some time (ten years or more) before this can be considered more than exploratory, assuming the technical requirements can be met economically.

An attractive idea is to remove the actinide alpha-emitters with the very long half-lives from the bulk of the wastes, especially from the high-level waste. The remaining fission product contaminated material then would need to be isolated up to only 1000 years, a more workable time period. Without the high heating generating fission products, the low heat generating actinides could be disposed of in some more suitable manner or recycled back to nuclear reactors for burning (transmutation) to shorter lived or stable elements. It is technically feasible to remove the actinides to negligible concentrations but uncertain if it is operationally obtainable. Furthermore, it will take several decades to demonstrate the feasibility of actinide recycle.

RADIOACTIVE WASTE MANAGEMENT OPERATIONS

Finally, it must not be overlooked that most radioactive waste treatment processes themselves generate intermediate- and low-level liquid wastes, mostly from process off-gas treatment, process condensates and decontamination solutions, and considerable volumes of contaminated equipment and trash. Such wastes are either recycled through the treatment processes, eventually resulting in a releasable effluent or added to quantities of radioactive waste to be stored of disposed of. Consequently from a waste management standpoint, the less radioactive wastes need to be handled, the better.