Radioactive waste management at nuclear power plants

An overview of the types of low- and intermediate-level wastes and how they are handled

by V. M. Efremenkov

In many countries nuclear power plants are an important part of the national energy system. Nuclear power is economically competitive and environmentally clean compared to most other forms of energy used in electricity production. Used in conjunction with them, it contributes to the security of national electricity supplies. It seems certain that in the medium term and beyond, a growing contribution to national energy supplies from nuclear energy will continue to be necessary if the standard of living in industrialized countries of the world is to be maintained and the energy needs of the developing countries are to be met.

As a result of the operation of nuclear reactors, some radioactive wastes are produced. Yet compared to the amount of waste produced by coal-fired electrical generating plants, these are of considerably smaller volume. *(See table.)* The wastes generated at nuclear power plants are rather low in activity and the radionuclides contained therein have a low radiotoxicity and usually a short half-life. However, nuclear power plants are the largest in number among all nuclear facilities and produce the greatest volume of radioactive wastes.

The nature and amounts of wastes produced in a nuclear power plant depend on the type of reactor, its specific design features, its operating conditions and on the fuel integrity. These radioactive wastes contain activated radionuclides from structural, moderator, and coolant materials; corrosion products; and fission product contamination arising from the fuel. The methods applied for the treatment and conditioning of waste generated at nuclear power plants now have reached a high degree of effectivity and reliability and are being further developed to improve safety and economy of the whole waste management system.

Wastes generated at nuclear power plants

Low- and intermediate-level radioactive waste (LILW) at nuclear power plants is produced by contamination of various materials with the radionuclides generated by fission and activation in the reactor or released from the fuel or cladding surfaces. The radionuclides are primarily released and collected in the reactor coolant system and, to a lesser extent, in the spent fuel storage pool.

The main wastes arising during the operation of a nuclear power plant are components which are removed during refuelling or maintenance (mainly activated solids, e.g. stainless steel containing cobalt-60 and nickel-63) or operational wastes such as radioactive liquids, filters, and ion-exchange resins which are contaminated with fission products from circuits containing liquid coolant.

In order to reduce the quantities of waste for interim storage and to minimize disposal cost, all countries are pursuing or intend to implement measures to reduce the volume of waste arisings where practicable. Volume reduction is particularly attractive for low-level waste which is generally of high volume but low radiation activity. Significant improvements can be made through administrative measures, e.g. replacement of paper towels by hot air driers, introduction of reusable long-lasting protective clothing, etc., and through general improvements of operational implementation or "housekeeping".

Liquid wastes and wet solid wastes

According to the different types of reactors now operating commercially all over the world, different waste streams arise. These streams are different both in activity content and in the amount of liquid waste generated. Reactors cooled and moderated by water generate more liquid waste than those cooled by gas. The volumes of liquid waste generated at boiling-water reactors (BWRs) are significantly higher than at pressurized-water reactors (PWRs). Because the cleanup system of heavy-water reactors (HWRs) works mainly with once-through ion-exchange techniques to recycle heavy water, virtually no liquid concentrates are generated at them.

Active liquid wastes are generated by the cleanup of primary coolants (PWR, BWR), cleanup of the spent fuel storage pond, drains, wash water, and leakage waters. Decontamination operations at reactors also

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generate liquid wastes resulting from maintenance activities on plant piping and equipment. Decontamination wastes can include crud (corrosion products) and a wide variety of organics, such as oxalic and citric acids.

Wet solids are another category of waste generated at nuclear power plants. They include different kinds of spent ion-exchange resins, filter media, and sludges. Spent resins constitute the most significant fraction of the wet solid waste produced at power reactors. Bead resins are used in deep demineralizers and are common in nuclear power plants. Powdered resins are seldom used in PWRs, but are commonly used in BWRs with pre-coated filter demineralizers. In many BWRs, a large source of powdered resin wastes are the "condensate polishers" used for additional cleaning of condensed water after evaporation of liquid wastes.

Pre-coated filters used at nuclear power plants to process liquid waste produce another type of wet solid waste-filter sludges. The filter aids — usually diatomaccous earth or cellulose fibres — and the crud that is removed from the liquid waste together form the filter sludges. Some filtration systems do not require filter aid materials. The sludges arising from such units therefore do not contain other materials.

Treatment and conditioning of liquid/solid waste

Liquid radioactive waste generated at nuclear power plants usually contains soluble and insoluble radioactive components (fission and corrosion products) and non-radioactive substances. The general objective of waste treatment methods is to decontaminate liquid waste to such an extent that the decontaminated bulk volume of aqueous waste can be either released to the environment or recycled. Waste concentrate is subject to further conditioning, storage, and disposal. Because nuclear power plants generate almost all categories of liquid waste, nearly all processes are applied to treat radioactive effluents. Standard techniques are routinely used to decontaminate liquid waste streams. Each process has a particular effect on the radioactive content of the liquid. The extent to which these are used in combination depends on the amount and source of contamination. Four main technical processes are available for treatment of liquid waste: evaporation; chemical precipitation/flocculation; solid-phase separation; and ion exchange.

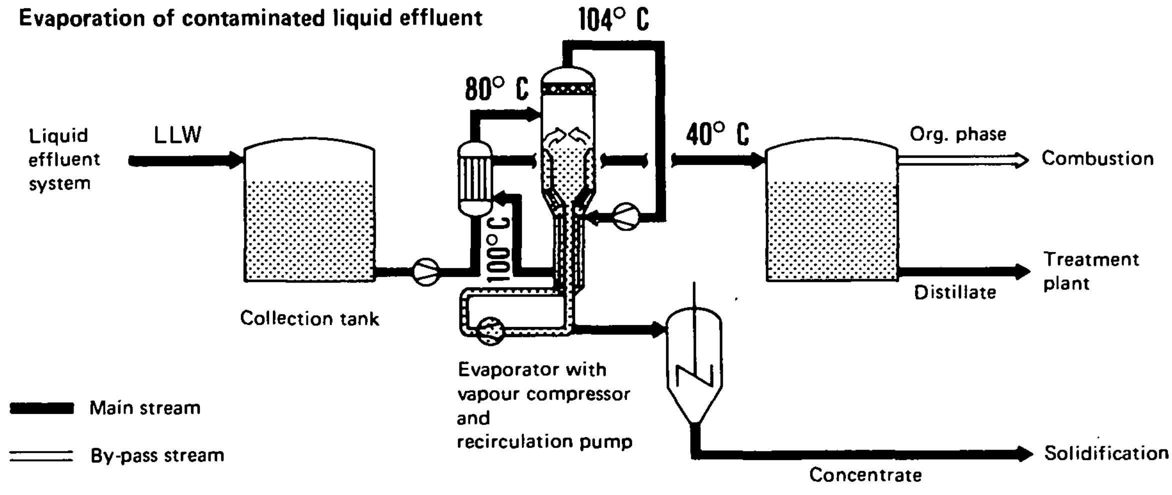
These treatment techniques are well established and widely used. Nevertheless, efforts to improve safety and economy on the basis of new technologies are under way in many countries.

The best volume reduction effect, compared with the other techniques, is achieved by evaporation. Depending on the composition of the liquid effluents and the types of evaporators, decontamination factors between 10^4 and 10^6 are obtained.

Evaporation is a proven method for the treatment of liquid radioactive waste providing both good decontamination and volume reduction. Water is removed in the vapour phase of the process leaving behind non-volatile components such as salts containing most radionuclides. Evaporation is probably the best technique for wastes having relatively high salt content with a wide heterogenous chemical composition. *(See accompanying figure.)*

Although it can be considered a fairly simple operation which has been successfully applied in the conventional chemical industry for many years, its application in the treatment of radioactive waste can give rise to some problems such as corrosion, scaling, or foaming. Such problems can be reduced by appropriate provisions. For example, the pH value can be adjusted to reduce corrosion; organics can be removed to reduce foaming or anti-foaming agents can be added; and the

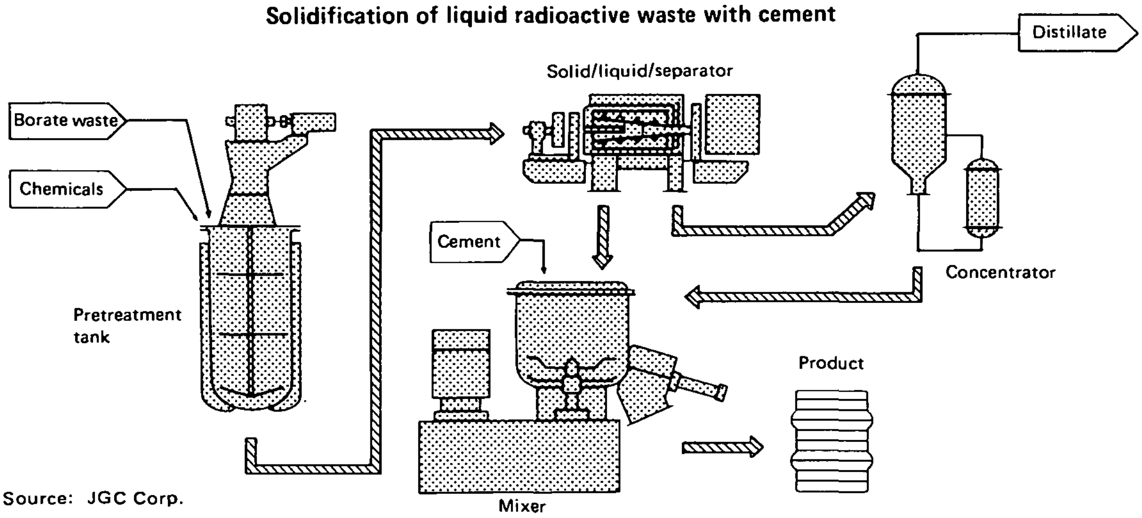
Evaporation of contaminated liquid effluent



Source: KFZ, Karlsruhe, Fed. Rep. of Germany.

LLW-Evaporation

Solidification of liquid radioactive waste with cement



Source: JGC Corp.

evaporator system can be cleaned by nitric acid to eliminate scaling and subsequent passivation of construction material.

Up till now, volume reduction by evaporation of low-level radioactive effluents has always been so effective that the clean condensate could be discharged to the environment without further treatment.

Chemical precipitation methods based on the coagulation-flocculation separation principle are mostly used in nuclear power plants for the treatment of liquid effluents with low activity and high salt and mud contents. Their effectiveness depends largely on the chemical and radiochemical composition of the liquid waste. Most radionuclides can be precipitated, co-precipitated, and adsorbed by insoluble compounds, e.g. hydroxides, carbonates, phosphates, and ferrocyanides, and so be removed from the solution. The precipitates also carry down suspended particles from the solution by physical entrainment. However, the separation is never complete for several reasons, and the decontamination factors achieved can be relatively low. For this reason, chemical treatment is usually used in combination with other more efficient methods.

Solid-phase separation is carried out to remove suspended and settled solid matters from the liquid waste. There are several types of separation equipment available, all based on those which have been regularly used in the conventional water and effluent treatment plants in the industries. The most popular types are filters, centrifuges, and hydrocyclones. Particle separation is a well-established technology. Almost all nuclear facilities use mechanical devices to separate suspended solids from liquid waste streams. Generally, separation equipment is needed to remove particles which could interfere with subsequent liquid waste treatment processes, e.g. ion exchange, or with the re-use of the water.

Typical filters can remove particles down to submicron sizes, particularly when a precoat is used. Once exhausted, the filter is either "backwashed" to yield a sludge of around 20–40% solids, or in the case of cartridge types, the entire unit is replaced.

Ion-exchange methods have extensive application in the treatment of liquid effluents at nuclear power plants. Examples of these include the cleanup of primary and secondary coolant circuits in water reactors, treatment of fuel storage pond water, and polishing of condensates after evaporation.

Liquid radioactive wastes usually have to satisfy the following criteria to be suitable for ion-exchange treatment: the concentration of suspended solids in the waste should be low; the waste should have low (usually less than 1 gram per litre) total salt content; and the radionuclides should be present in suitable ionic form. (Filters pre-coated with powdered resin can be used to remove colloids.)

In most technical systems, ion-exchange processes are applied using a fixed bed of ion-exchange material filled in a column which is passed through by the contaminated effluent either from top to bottom or vice-versa. The ion-exchange material may be regenerated after having reached saturation of the active groups (break through capacity). Some types of ion exchangers are also removed as waste concentrate to be solidified. Therefore the ion-exchange process represents a semi-continuous process and requires major efforts in maintenance like flushing, regeneration, rinsing, and refilling operations.

Wet solids resulting from liquid waste treatment must still be transformed into solid products for final disposal. Immobilization processes involve the conversion of the wastes to chemically and physically stable forms that reduce the potential for migration or dispersion of radionuclides by processes that could occur during storage, transport, and disposal. If possible, waste conditioning should also achieve a volume reduction.

The most frequently applied methods for conditioning wet solids are cementation, bituminization, or incorporation into polymers. Immobilization of radioactive waste using cement has been practised widely for many years in many countries. *(See accompanying figure.)* Cement has a number of advantages, notably its low cost and the use of relatively simple process plant. Its relatively high density provides the waste forms with a considerable degree of self-shielding thereby reducing requirements for additional package shielding. In certain cases, in order to achieve a product of acceptable quality, chemical or physical pre-treatment steps may be employed. Sometimes additional alternative materials, such as pulverized fuel ash and blast furnace slag, can be used. These behave in a similar way to simple cement.

Bitumenization also has been used for a number of years in various countries for solidification of wet solids. Bitumenization is a hot process which allows the wet stream to be dried off before being immobilized and packaged. This greatly reduces the volume of conditioned waste requiring disposal with a consequent saving in cost. However, bitumen is potentially flammable requiring special precautions to prevent its accidental ignition. Nevertheless, bitumenization has found growing acceptance with waste producers and is used for conditioning of radioactive waste at nuclear power plants in the USA, Japan, Sweden, USSR, Switzerland, and other countries.

Incorporation of wet solids into plastics or polymers is a relatively new immobilization process when compared to the use of cement or bitumen. The use of polymers such as polyester, vinylester, or epoxide resins is generally limited to those applications where cement or bitumen are technically unsuitable. Such polymers are considerably more expensive and a relatively complex processing plant is needed. Polymers have the advantages of offering greater leak resistance to radionuclides and of being generally chemically inert.

There has recently been increased interest in the use of mobile units to condition radioactive waste from nuclear power plants. This has arisen mainly because they provide saving in capital cost where on-site arisings of waste are small. Mobile immobilization units for conditioning of radioactive waste of nuclear power plants are used, for example, in the USA, Federal Republic of Germany, and France. Most of them utilize the cementation process, although several designs for utilizing polymers have been developed.

Gaseous waste and radioactive aerosols

In normal operation of nuclear power plants, some airborne radioactive wastes are generated in either particulate or aerosol of gaseous form. Particulate radioactive aerosols can be generated in a wide range of particle sizes in either liquid or solid form, possibly in combination with non-radioactive aerosols. Three main sources of aerosols are generated by emission of activated corrosion products and fission products; radioactive decay of gases to involatile elements; and adsorption of volatile radionuclides formed in the fission process on existing suspended material.

The most important volatile radionuclides, which form gaseous radioactive waste generated during normal operation of nuclear power plants, are halogens, noble gases, tritium, and carbon-14. The composition and the amount of radioactivity present in the various airborne waste streams largely depend on the reactor type and the release pathway.

All gaseous effluents at nuclear power plants are treated before discharge to the atmosphere to remove most of the radioactive components from the effluence.

Treatment of gaseous effluents

It is common practice at all nuclear power plants for contaminated gases and building ventilation air to be first passed through filters to remove particulate activity before discharge to the atmosphere via stacks. Ventilation and air cleaning system usually employ coarse pre-filters followed by high-efficiency-particulate-air (HEPA) filters. These have typical particle removal efficiencies of 99.9% or better for 0.3 mm particles.

Radioactive iodine arising from power plant operation is routinely removed by impregnated charcoal filters, used in combination with particulate filters. Impregnation is required to trap the organic iodine compounds from gas effluents.

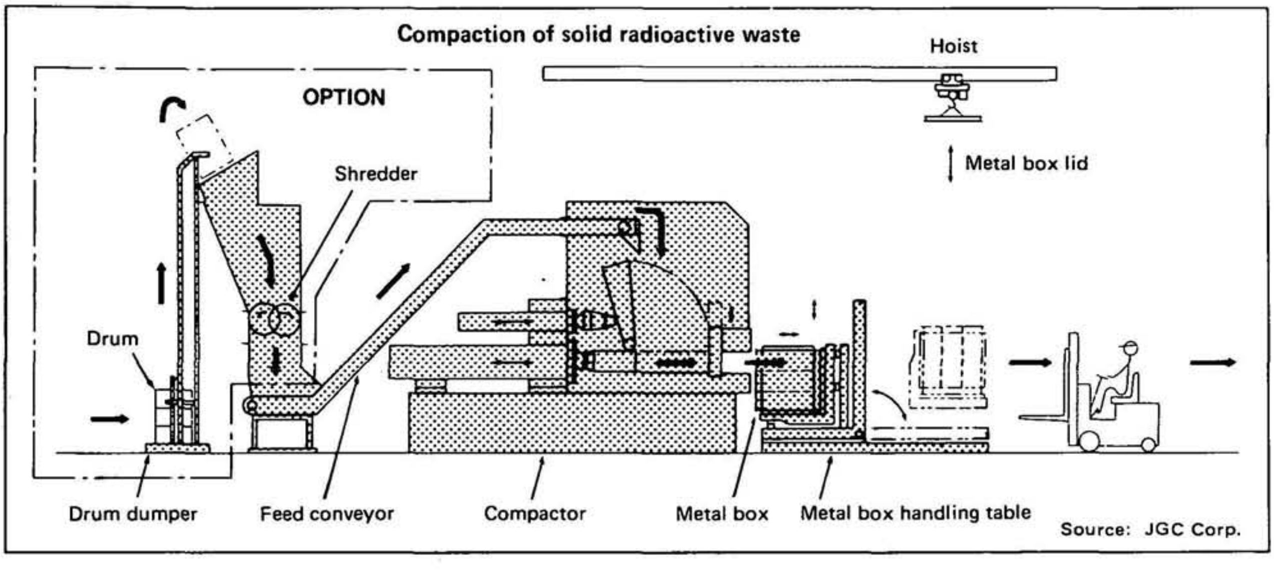
Because noble radioactive gases released from fuel elements in a small amount are mainly short-lived, delaying their release will allow radioactive decay processes to greatly reduce the quantities finally released to the environment. Two delay techniques are used for this purpose: storage in special tanks or passage through charcoal delay beds.

For decay storage, the noble gases and their carrier gas are first pumped into gas tanks which are then sealed. After a storage time between 30 and 60 days, the content of the tanks is ventilated to the atmosphere through a ventilation system. If release is not permissible, the storage period is extended as necessary.

Delay beds consist of a number of vessels filled with charcoal, which relatively retards the passage of noble gases in relation to the carrier gas and allows radioactive decay to take effect.

Treatment and conditioning of solid waste

During the operation of a nuclear power plant, various types of dry solid wastes containing radioactive materials are generated. The nature of these wastes vary considerably from facility to facility and can include redundant items of the reactor plant, ventilation system filters, floor coverings, contaminated tools, etc. Another source of solid waste is the accumulation of miscellaneous paper, plastic, rubber, rags, clothing, small metallic or glass objects, used during the operation and maintenance of the nuclear power plant. Depending on the



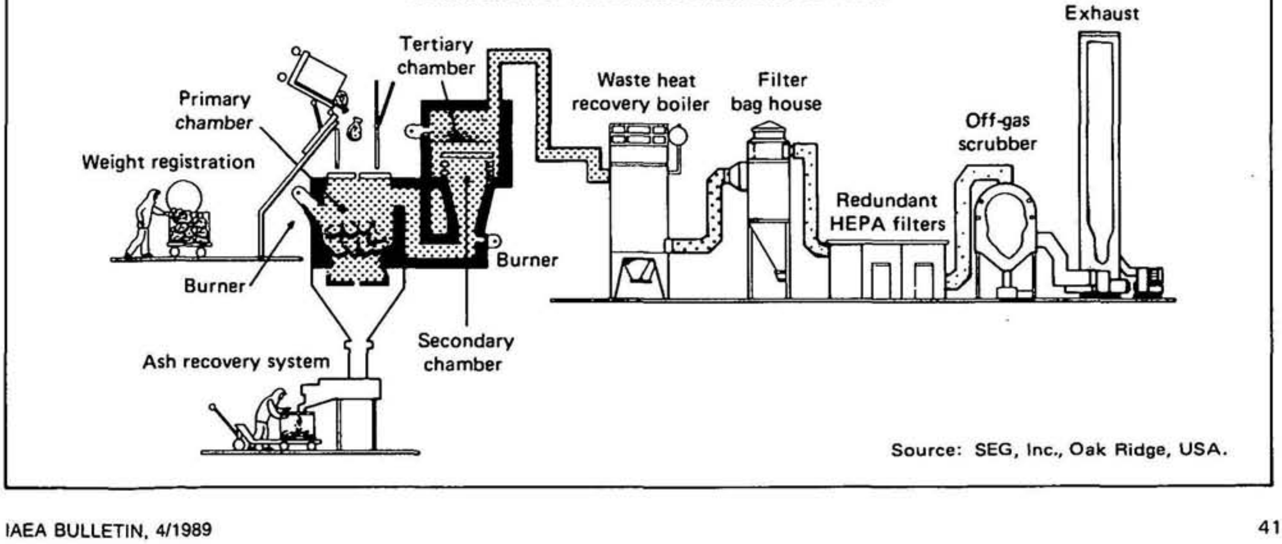
physical nature and further treatment methods, dry solid waste usually is classified and segregated into four main categories: combustible, non-combustible, compactible, and non-compactible waste. However, each facility usually has its own level of classification according to the prevailing conditions.

One of the essential aims in the treatment of solid waste is to reduce as much as possible the waste volumes to be stored and disposed of and to concentrate and immobilize as much as possible the radioactivity contained in the waste.

As solid radioactive waste at nuclear power plants consists of a broad spectrum of materials and forms, no single technique can adequately treat this waste; a combination of processing techniques is generally used. The basic and most common technique used for processing most voluminous portions of solid waste has been based on compaction. This method reduces the storage and disposal volume requirements by a reasonable amount, but achieves little in terms of improvement of the waste properties from the viewpoint of longer term management.

Experience has shown that between 50% and 80% of solid radioactive waste produced at nuclear power plants can be classified as burnable waste. Incineration of this waste represents a substantial improvement from a number of viewpoints over simple compaction. Very high volume reduction and mass reduction can be achieved. The final product is an homogeneous ash which can be packaged without further conditioning into containers for storage and disposal. While incineration is only suitable for combustible waste, it has the advantage of being capable of destroying organic liquids, e.g. oils, greases or solvents, which otherwise are difficult to treat. *(See accompanying figures.)*

Incineration of small quantities of solid waste is routinely carried out in relatively simple units. Such incineration facilities have now been installed at the



nuclear power plants in the USA, Japan, Canada, and other countries. More advanced incineration facilities that can incinerate waste with relatively high specific activity are installed at centralized waste treatment facilities that can accept waste from many plants in the country and from abroad. Such facilities are operating in Sweden, Belgium, France, and other countries.

As a pretreatment step for compaction or incineration, cutting, shredding, and crushing are used to reduce the physical size of individual waste items. Paper, plastic, cloth, cardboard, wood, and metals can be shredded into ribbon-like pieces, while brittle materials such as glass or concrete blocks can be crushed into smaller fragments. These techniques can also be used as stand-alone processes for volume reduction of solid waste.

New developments

Most treatment and conditioning processes for LILW have now reached an advanced industrial scale. Although these processes and technologies are sufficient for effective management of radioactive waste at nuclear power plants, further improvements in this technology are still possible and desirable. The increasing cost of radioactive waste disposal provides an incentive to adopt procedures and techniques to minimize waste quantities and to develop new techniques to minimize volumes at the treatment and conditioning step. It is not possible here to summarize all new developments and improvements which are being made in this direction in Member States.

Some examples of such new developments include the use of specific inorganic sorbents to improve liquid waste treatment; use of membrane techniques for liquid waste treatment; dewatering and drying of bead resin and filters slurries; incineration of spent ion-exchange resins; dry cleaning of protective cloth to reduce quantity of laundry drains; use of high integrity containers for packaged dried filter sludges; vitrification of some intermediate-level waste to reduce volumes of waste to be disposed of; and supercompaction of unburnable waste.

Perhaps not all of these new developments will find broad implementation in waste management technologies, in particular at nuclear power plants. However, the research and development reflects the fact that the nuclear industry and utilities take great care in a safe and economic management of radioactive waste at nuclear power plants, and that improvements in existing technology are forseen.

At the Marcoule facility in France, containers of vitrified high-level waste are transferred to air-cooled shafts for interim storage. (Credit: ANDRA)