WASTE MANAGEMENT EXPERIENCE AT IPEN - BRAZIL

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ABSTRACT

This paper describes the history and the practices of the waste management at a nuclear research center in Brazil where research on the nuclear fuel cycle and on the applications of radioisotopes are in progress.

INTRODUCTION

The Institute for Energy and Nuclear Research (Instituto de Pesquisas Energéticas e Nucleares - IPEN) is the biggest nuclear research center in Brazil. Located in the campus of the University of São Paulo, it is maintained and operated by the National Commission on Nuclear Energy (Comissão Nacional de Energia Nuclear - CNEN). Its objectives are the development of nuclear energy and its fuel cycle, the applications of radiation and radioisotopes in industry, medicine, agriculture, research, education, and environmental preservation, and the realization of basic and applied research in related fields.

The history of IPEN (formerly Institute of Atomic Energy - IEA) begun in 1957 when the 2 MW swimming pool reactor IEA-R1 started operation. IPEN are producing radioisotopes for medical and industrial applications since 1961.

Presently, installations at IPEN include many facilities of the nuclear fuel cycle, two reactors, facilities for the production of radioisotopes and labeled compounds, facilities for the production of radioactive sealed sources for industrial applications and laboratories for research on nuclear physics, radiochemistry, radiobiology and nuclear medicine.

Radioactive wastes arising from those facilities range widely in form, activity, and radionuclide content. Quantities are typically in the range of 40 to 200 m^3 per year and activities vary from the few MBq/m^3 to the several GBq/m^3. Waste types can be classified for management purposes as: solids, wet solids, and liquids.

The wastes with the highest activities arise in the production of radioisotopes although they are contaminated with short lived radionuclides. Uranium and thorium refining facilities generate low level wastes but contain the most radiotoxic and long-lived radionuclides.

The users of radioisotope and sealed sources also send their wastes to IPEN for treatment. This waste stream, specially spent sealed sources, represents a significant fraction of waste volumes being handled.

The history of waste management at IPEN can be divided in two periods: one spanning from its beginnings in 1957 until 1979, when waste management practices consisted almost exclusively of the burial of all solid wastes and the decay/delay/discharge scheme for all liquid wastes; and the second period, that is about to end now, when volumes and activities increased and a wider range of waste types required more technology, budget and trained personnel for their management. It started when CNEN prohibited waste burial at IPEN, what leads to the project and construction of the facilities for compaction, encapsulation and interim storage of wastes that are in operation since the middle of 1982.

As many as twenty professionals, ten of them with academic degree, worked in routine as well as in research and development activities during this time, by contrast to the previous period when a team of two or three technicians from the Health Physics Division collected and treated the wastes at IPEN.

The ocurrence of the radiological accident in Goiânia in the end of 1987 is also noteworthy in this period because this event directly and indirectly contributed to a significant increase in waste volumes being handled. As a direct consequence of the accident, almost 85 m^3 of metal scrap and paper bundles contaminated with ^{137}Cs, found in cities near São Paulo, were collected and transported to IPEN. Indirectly the accident caused an increase in waste volumes because of a more stringent control over radioactive sources and radioisotope users all over the country. As a result, since the time of the accident, increasing quantities of spent sources and other wastes reached IPEN for proper treatment.

The history of waste management at IPEN is now starting its third period. Waste management staffs are now facing problems of budget cuts because of uncertainties in the future of nuclear energy in Brazil, besides coping with the challenge of dealing with wastes of higher activities that will arise from a fission-^{99}Mo production facility under project.

The following sections have a description of the facilities, experience and achievements, an appraisal of the present status, and an evaluation of the near future scenario.

WASTE MANAGEMENT AT IPEN

The wastes managed at IPEN are of low activity and can be classified as:

solids - compressible low-level laboratory wastes; non-compressible metal scrap and laboratory decommissioning wastes; biological solid wastes, mainly guinea pigs and rats;

wet solids - ion exchange resins; activated charcoal filter beds; slurries and cakes from uranium and thorium refining plants;

liquids - scintillation cock-tails; organic solvents; aqueous efluents from radioisotope production facilities and radiochemistry laboratories;

The main problem to be solved initially was the destination of contaminated compressible laboratory trash which amounted yearly to some tens of cubic meters and that no longer could be buried as previously because the shallow ground burial of solid wastes was forbidden by decision of CNEN.

Consequently volume reduction equipment was necessary for the waste to be stored before transportation to a disposal site. Due to the nature of the wastes, the choice fell on a ten ton hydraulic bailing press adapted to compact wastes directly inside 200 L drums.

A room, about one hundred square meters, was built to house the compaction unit. The building was provided with a ventillation system, floor drain with an underground tank, a shielded area to store the waste before compaction, radiation monitors, and a glove box for cutting and dismantling pieces do not fit inside the 200 litre drums. A filtration system with HEPA filter and an activated charcoal bed with 95% iodine adsorption efficiency was installed in the ventillation system. An epoxide resin was used to cover floor surface and walls up to 50 cm high. The drain system directs floor decontamination effluents to an underground retention concrete tank with high liquid level alarm and means to pump the liquid to the sewer or to a mobile tank.

Tin plated #18 US gauge steel drums are used because thinner drum walls usually may be perforated by rigid pieces of material inadvertently put together with compactible waste or because it may be distorted by the thrust of the press over the waste.

In ten years of operation as many as 500 cubic meters of compactible solid waste were compacted to a volume four to five times lower, resulting in about 550 drums.

Some non-compressible wastes are produced in the laboratories of IPEN and the treatment for them is the simple packaging in drums or sometimes the encapsulation in concrete. The encapsulation takes place when exposure rate at drum surface exceeds transportation limits or when a postulated acident with the drum during handling or transportation is deemed to cause significant contamination. The concrete is prepared in a standard concrete mixer and transferred to the drum while a needle type vibrator is used to ensure a dense and voidless mass among waste pieces. About 170 drums filled with noncompressible waste are stored presently.

Spent radioactive sources fall in this class of waste. Industrial as well as many radiotherapy sources sent to IPEN for treatment as waste are encapsulated in concrete. Most sources comming from industry have their shieldings disassembled for reuse and are put in lead castles. A number of castles are encapsulated in one drum so as to not exceed the limits of activity and exposure rate set forth by the transportation standards while using the maximum of drum capacity. Damaged shieldings that can not be safely disassembled or sources with activity near the transportation limits are encapsulated in the drum without removal of the source from the original shielding. As the maximum permissible activity per package it is considered the limit for type A1 package of the Safety Series No. 6.

Thirteen drums are filled with encapsulated spent sources.

The exception in this procedure is the encapsulation of a leaking ^{226}Ra-Be neutron source, for which a special package must be used. Besides the necessity of neutron shielding, the require-ments for the encapsulation of radium sources are very stringent. The necessary shielding thickness for the neutrons and gammas was evaluated using proper softwares. It was concluded that Ra-Be sources with less than about 3 GBq can be encapsulated without any additional neutron shielding material in the package usually used for disposal of radium sources. This package will be described later.

From hospitals some teletherapy ^{60}Co sources and hundreds of ^{226}Ra needles are being collected. Cobalt sources because of decay to activities lower than practical values for therapy and radium needles because of changes in practices.

These sources are more dificult to manage. The cobalt sources can not be encapsulated because they are too intense to be handled in our installations, and so efforts are made for the source to be reused in other aplications. The problems with radium needles are related to the specifications of packages. CNEN imposed very stringent requirements on the packages: very low leak rates from the containment of the sources, high quality container and use of adsorbers inside the package for retention of radon. The allowable leak rate is 10^{-6} torr L s^{-1} measured with a helium leak detector. Adsorbers shall be activated charcoal, not less than 1 g of charcoal per mCi of radium, (formerly 10 g charcoal per mCi radium inside the containment). Containment and package materials should be stable materials with high durability. The package is therefore composed of a stainless steel capsule, 1.6 litres capacity, closed by a lid held in position by six screws and sealed with a soft copper disk gasket. Shielding is made up of a lead sheet, 3 mm thick, around the capsule, a precast barite concrete layer inside a drum, and a fresh barite concrete seal. The 200 litres standard steel drum made up the external package. Up to 17 GBq (500 mCi) of ^{226}Ra is allowed in each drum corresponding to 50 to 100 needles per package. Up to now, 17 drums containing radium needles were produced.

Another solid waste requiring management now is the radioactive lightning rods discarded as waste, after a resolution of CNEN determined that new lightning rods containing radioactive sources should not be installed and that all those requiring maintenance should be replaced by conventional Franklin type. The radioactive ones should be sent to CNEN for proper treatment. There is an estimate of over 80.000 such rods to be treated. About 5,000 lightning rods were received to date and had to be dismantled for removing the americium sources from the bulky metal structure. The main problem is that many of the americium sources are leaking because, although covered with a gold layer that provides a chemically resitant surface, the source is not safe against wind borne dust erosion. Americium-241 is one of the most radiotoxic radionuclides and the inhallation of a single aerosol particle of pure americium or of dust with attached americium a few tens of micrometers across may account for the activity of the Annual Limit on Intake (ALI). The sources in each lightning rod have together about one million ALIs. So, the dismantling operations are done in a ventilated fumehood with aerosol filters to protect workers and to prevent air contamination.

Liquid wastes from radioisotope production are usually stored for decay in glass bottles at the plant where they were produced, to avoid the risks of transportation. When decayed they are mixed with other laboratory efluents, neutralized and discharged. The waste from the production of ^{131}I is one exception. Radioiodine is produced by irradiation of natural tellurium and the wet distillation of iodine gives rise to a 6 molar sulfuric acid solution with dissolved tellurium isotopes, mainly ^{123m}Te (T1/2 = 120 days), ^{125m}Te (T1/2 = 57 days), ^{127m}Te (T1/2 =109 days). The volume is about 2 litres per production shift (2 or 4 L per week) and activity at the time it is collected is on the order of tens of GigaBecquerels (a few Curies). The decay time to allow discharge to the sewer is about ten years. The storage in liquid form is hazardous and some incidents give rise to contamination of installations and some times air contamination with the residual iodine volatilized by the oxidizing sulfuric media.

A process is being developed in which the waste is neutralized stoichiometricaly with calcium hydroxide to form calcium sulfate that sets as common gypsum yielding a dry, solid material. It allows the safe storage of the waste in drums without any noticeable corrosion.

Many other miscellaneous small volume liquid wastes are routinely generated in radiochemical laboratories and are all being subject to decay/delay/discharge management schemes.

Liquid wastes generated by radioisotope users are seldom sent to IPEN for treatment because most liquid wastes are contaminated with short-lived radionuclides. So, radioisotope users manage their wastes themselves, frequently dis-charging them directly on the sewer and some times allowing them to decay before discharge. Liquid wastes are sent to IPEN either when activity levels exceed discharge limits and half-lives are long or when the customer has no means to determine activity concentration. In these cases wastes are accepted and the activity is measured and the waste is duly treated.

IPEN received during some time scintillation cocktails contaminated with ^{3}H or ^{14}C from diagnostic clinics, although the activity levels were lower than the limits. IPEN received annualy about 1.5 m^3 of cocktails and treated them by distillation to reduce volume, reduce chemical pollution and to recycle toluene, used as solvent in the cocktails. The bottoms of the distillator were later discharged to the sewer and the recycled toluene was reused in cocktail preparation. Counting in liquid scintillation counter showed that the recycled toluene was decontaminated and infrared spectroscopy showed it was free from organic impurities. Nevertheless, this practice was stopped because of unbalance in expenditures and income.

Other wastes currently being handled are wet or damp solids from pilot scale uranium conversion facility and ion exchange resins and activated charcoal from the research reactor coolant polishing system. The wet waste coming from uranium conversion is a silica cake made of the insoluble matter that ramains in the filter after dissolution and filtration of the yellow cake. It contains about 78 % water, 20 % silica, 0.3 % iron, 0.1 uranium, some anions, mainly nitrate, and many other minor metal cations. It is contaminated with natural uranium, ^{230}Th and ^{226}Ra and is suspected to contain other long lived radionuclides of the uranium and actinium series such as ^{227}Ac, ^{231}Pa and ^{210}Pb. On average, nitric acid is present in 0.4 meq per gram of cake. A complete chemical and radiological characterization of this waste is in progress.

Ion exchange resins and activated charcoal are contaminated with activation products, mainly ^{60}Co, and fission products that leak from the fuel elements. The activation products come not only from the core structure but also from targets irradiated in the reactor for the production of industrial sources and for research. They are stored and will be immobilized in cement in the near future.

For the treatment of these wet solid wastes an immobilization system was built. It is composed of a mixer adapted to act as an 'in drum' planetary mixer and a cement feeding sistem. Its is suitable for low activity waste and is used primarily to immobilize the silica cake and as a mockup of a cementation unit that is under project. The silica cake is solidified in cement in a 0.8 waste/cement ratio and final product has no free water. The mechanical strength of the waste product is about 8 MPa, as determined by axial compression test.

Interim storage of waste drums was initially done over concrete platforms where the drums were stacked and covered with canvas. Later the platforms were replaced by 2 sheds, 200 m^2 each one, which afford greater protection against weathering. Over 900 drums are stored today.

PRESENT SITUATION

The volume of wastes generated at IPEN is decreasing because of the deep cuts in investments in the nuclear activities that are occuring in the last years.

However, the amount of waste reaching IPEN from abroad for treatment increased sharply after the accident in Goiânia at the end of 1987. The accident had impacted waste management activities in three different ways: a) wastes generated directly by the decontamination operations; b) wastes generated by the psychological impact of the accident on general public that exerted pressure over radioisotope users; c) wastes generated by the increased control over radioisotope users and changes in regulations.

a) Some time before the accident had been noticed two weeks after the cesium capsule had been opened, about 50 paper bundles, 400 kg each, were transported by truck to some Sao Paulo state cities where they would be reprocessed. The contaminated paper bundles were all rescued but the decontamination of the trucks and plants generated aditionally one hundred drum of soil, paving bricks, metal scrap and decontamination materials.

The medical care of some irradiated patients in Rio de Janeiro generated a few tens of drums with compacted or cemented waste that were also stored at IPEN.

All these accident related wastes are waiting transport back to Goiania where they will be disposed of together with the 4,000 cubic meters of wastes generated in the decontamination of Goiânia.

b) The curve in the graph of the annual waste volumes arising at IPEN from radioisotope users and mainly from industry, has a marked increase during and after 1988. The reason is linked to the impact the accident had on general public. To ilustrate what is stil happening it is claimed that the alleged reason for a plant manager to dispose of a complete and yet operating instrument that uses a small radioactive source gauge was: "We are replacing the control equipment by one that does not use a 'sensible' technology", 'sensible technology' meaning the one that uses radioactive material. Another user alleged pressure to remove the source from the 'working area'.

All these occurred as consequence of the fear the accident left on people and the lack of a policy to increase the public understanding and confidence in the applications of radioactive sources.

c) Last, the accident showed the necessity of a closer control over radioactive material users and in consequence some source owners decided to discontinue the practice instead of adhering to the more stringent requirements of CNEN. This resulted in more sources being discarded as waste.

FUTURE PROSPECTS

The amount of waste, mainly spent sources, coming from users is likely to decrease because the impact of the Goiânia accident is no long felt and the situation is nearing stability.

It is expected also that government will resume investments in nuclear industry in the near future which will push figures upward.

It is expected that intermediate level wastes will be generated in the near future. A facility for the production of ^{99}Mo is being designed to produce about 4 TBq (100 Ci) per week. It is calculated that about 100 TBq (2,700 Ci) of fission products will remain weekly in liquid and solid wastes at that plant. To manage this level of activity significant effort is necessary because of the lack of previous experience. However a concept of the facility for waste treatment and immobilization in cement is already complete and the characterization of product as well the characterization of raw waste is in progress.

The design of the process and hot cells to treat and to immobilize the several tens of TeraBecquerels of fission products generated thereupon is the job for the coming years. This mission has triggered research work in waste characterization and immobilization, waste form characterization, package design, hot cell design, process development, etc.