TREATMENT AND CONDITIONING OF INSTITUTIONAL WASTES IN BRAZIL

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Abstract

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An important condition for the expansion of electricity generation by nuclear reactors in the near future is improved control of the nuclear fuel cycle. An important part of this cycle is the management of radioactive wastes. Brazil has one nuclear power plant in operation (626 MW(e)); a uranium mining and milling industry; a nuclear fuel fabrication plant; three nuclear research institutes, one of which includes a radioisotope production facility; several particle accelerators; and about two thousand institutions (in medicine, industry and agriculture, research centres, etc.) throughout the country utilizing radionuclides on a large scale. In Brazil a final repository to receive the radioactive wastes produced in the country has not yet been defined. Instead, all radioactive wastes produced must be treated in such a way that the waste volume is minimized as much as possible, the release of radionuclides into the environment is as low as possible, and the radioactive materials are immobilized for safe storage over long periods to prevent them from entering the biosphere. The paper discusses the Brazilian nuclear power policy and the waste treatment research programme carried out by the Instituto de Pesquisas Energéticas e Nucleares, the scientific branch of the Comissão Nacional de Energia Nuclear.

1. INTRODUCTION

Production of radioisotopes for medical and industrial purposes, and experiments in reactor physics, nuclear physics, etc., for personnel training were initiated in Brazil in the mid-1950s using the experimental reactor IEAR-1-5MW located in Saõ Paulo.

Small amounts of wastes have been produced since then and so far have been kept under radiological control. With the introduction of the Brazilian nuclear power programme and an increase in the use of radioisotopes in industry, medicine and other fields, and with growing interest in protecting public health and ensuring public

TABLE I. SEALED SOURCES USED IN INDUSTRY, RESEARCH AND MEDICINE RECEIVED BY THE IPEN

Source	Source strength (GBq)	Half-life (a)	Principal applications
H-3	1–1000	12.3	Production of neutrons by (D,T) reactions
Co-60	$\leq 4 \times 10^3$	5.3	Radiotherapy, industrial radiography, level gauge
Kr-85	0.1–50	10.7	Paper thickness measurement
Sr-90	0.1–2	28.6	Paper thickness measurement, beta therapy
Cs-137	$\leq 4 \times 10^3$	30.2	Radiotherapy, industrial radiography, density gauge, level gauge
Pm-147	1–10	2.6	Paper thickness measurement
Am-241	1–10	432	Density gauge, lightning rod
Am-241-Be	0.1–50	432	Moisture detector, hygrometer, petroleum detector

safety, a regulation was established covering all aspects of the use of radioactive materials in the nuclear fuel cycle as well as in nuclear activities resulting from the non-nuclear fuel cycle.

For wastes from the nuclear fuel cycle great emphasis is given to volume reduction, while for wastes from industry, medicine and research outside the nuclear fuel cycle volume reduction is not critical because the volumes generated are so small that they do not justify investment in a volume reduction facility, although they may be sent for treatment in institutions which already possess such facilities.

In Brazil the main sources of radwastes generate low level wastes (LLW) and only in a few cases intermediate level wastes (ILW). The wastes mainly originate from: one nuclear power plant in operation (626 MW(e)); the uranium mining and milling industry; a nuclear fuel fabrication plant; three nuclear research institutes, one of which includes a radioisotope production facility; several particle accelerators; and about two thousand establishments (in medicine, industry and agriculture, research centres, etc.) throughout the country.

Since at present there is no permanent site to receive the radioactive wastes produced in the country, the LLW and ILW must be treated and stored appropriately, under the supervision of the Comissão Nacional de Energia Nuclear (CNEN), by the producers or sent to the Instituto de Pesquisas Energéticas e Nucleares (IPEN), which is in charge of receiving wastes when the radioisotope producer is not able to handle them (Table I).

This paper discusses the Brazilian nuclear power policy and the research and development programme carried out by the IPEN, as well as plans for final disposal in the near future.

2. CLASSIFICATION OF WASTES

The official classification of wastes in Brazil originates from the standards laid down by the International Atomic Energy Agency (IAEA) and is entitled Gerência de Rejeitos Radioativos em Instalações Radioativas (CNEN-NE-6.05-Nov/1985).

TABLE II. CLASSIFICATION OF RADIOACTIVE WASTES IN BRAZIL

Category	Beta/gamma emitters (α emission $\leq 3.7 \times 10^8$ Bq/m^3)	Alpha emitters (α emission $> 3.7 \times 10^8$ Bq/m^3)
	Surface exposure rate, $\dot{X}$ (μC·kg^{-1}·h^{-1})	Concentration, c (Bq/m^3)
I. Solid wastes		
Low level	$\dot{X} \leq 50$	$3.7 \times 10^8 < c \leq 3.7 \times 10^{11}$
Intermediate level	$50 < \dot{X} \leq 500$	$3.7 \times 10^{11} < c \leq 3.7 \times 10^{13}$
High level	$\dot{X} > 500$	$c > 3.7 \times 10^{13}$
	Concentration, c (Bq/m^3)	
II. Liquid wastes		
Low level	$c \leq 3.7 \times 10^{10}$	$3.7 \times 10^8 < c \leq 3.7 \times 10^{10}$
Intermediate level	$3.7 \times 10^{10} < c \leq 3.7 \times 10^{13}$	$3.7 \times 10^{10} < c \leq 3.7 \times 10^{13}$
High level	$c > 3.7 \times 10^{13}$	$c > 3.7 \times 10^{13}$
III. Gaseous wastes		
Low level	$c \leq 3.7$	
Intermediate level	$3.7 < c \leq 3.7 \times 10^4$	
High level	$c > 3.7 \times 10^4$	

The classification is based on health and safety requirements established according to practical experience at waste treatment plants in other countries and applies to existing national and international regulations for the safe transport of radioactive materials.

The wastes are classified according to their physical state, the nature of the radiation, radionuclide concentration and exposure rate. Primarily the wastes are classified on the basis of their physical state, i.e. liquid, solid or gaseous. Another very important distinction is the presence or absence of alpha emitters in the waste stream. On the basis of this classification a more appropriate treatment method can be assigned. In addition, there is a more detailed classification on the basis of the radiological properties of the wastes. For liquid and gaseous wastes this classification is made according to specific activity while for solid wastes the classification is based on the exposure rate. The classification used in Brazil for solid, liquid and gaseous wastes according to their radiation contents is presented in Table II.

The elimination of certain wastes from any installation is subject to special authorization from the CNEN. The upper limit for the specific activity of solid wastes permitted to be disposed of in the urban refuse collection system is 74 Bq/g (2 nCi/g), while for liquid and gaseous wastes allowed to be eliminated in sewage or the atmosphere a table containing this information for each radionuclide is provided in the regulation mentioned at the beginning of this section.

According to this regulation persons engaged in activities involving radioactive materials must provide complete information and technical data on training, experience, safety procedures, the interim storage system and disposal of effluents, as well as a detailed analysis of the proposed activity.

A person applying to use radioisotopes may request the CNEN for specific approval of a proposed procedure to dispose of licensed radioactive materials in a manner other than that generally authorized in the regulation. If necessary, meetings are held between the CNEN staff and the applicant to solve specific questions.

To assure compliance with the approved conditions all licensees are subject to periodic inspection. Radioactive wastes which can no longer remain under the responsibility of the applicant are collected by the CNEN inspectors and sent to the IPEN for necessary treatment and intermediate storage.

3. RADIOACTIVE WASTES IN BRAZIL: GENERAL SITUATION

3.1. Wastes from institutions (medicine, industry, agriculture, etc.)

The wastes arising from the application of radioisotopes in the medical and biochemical research fields as well as in clinical medicine, together with the wastes resulting from the application of radioactive materials in industrial processes, are treated as follows:

- For short lived radionuclides such as ^{125}I, ^{131}I, ^{67}Ga, ^{201}Tl, ^{32}P, ^{35}S and $^{99}Tc^{m}$, the majority of the wastes from medical and research institutions are subjected to a decay treatment method followed by disposal into the sewage system.
- Solid wastes contaminated by traces of short lived radioisotopes with a specific activity not exceeding 74 Bq/g (2 nCi/g) can be sent to a municipal refuse disposal plant.
- The excreta of patients who have received therapeutic doses in excess of 555 MBq (15 mCi) have to be collected in bags and, after decay, emptied into a special sink.
- Radiotherapy sources, including sources used in sterilization, must be returned to the country of origin.
- Sources generally used in industrial equipment, such as for foil thickness measurement, level detection, gauging, quality control and smoke detection, are generally sent to the IPEN for treatment.
- Exhausted ^{192}Ir sources, mostly used in non-destructive testing, are also sent to the IPEN for recovery.

3.2. Wastes from research institutes

The principal waste forms generated by the research institutes are miscellaneous liquids, trash, biological wastes, scintillation vials and sealed sources and targets. Miscellaneous liquids containing short lived isotopes or small concentrations of radionuclides are generally released to the sewer. Trash, sealed sources and targets are normally packaged in 100 or 200 L drums. The IPEN gives special attention to the users of ^{3}H and ^{14}C contained in liquid scintillation vials and some animal carcasses, in order to ensure compliance with the regulations on radiation protection, transport and management of radwastes.

3.3. Wastes from the Brazilian nuclear power plant Angra I

The operation of nuclear power plants does not produce excessive amounts of wastes when considered in relation to the total fuel cycle; most of the wastes produced by them fall exclusively in the low and intermediate level waste categories. In spite of this in Brazil the nuclear power plant Angra I is the most important waste source. Table III indicates the approximate volumes of radioactive wastes generated by Angra I up to the end of 1987.

3.4. Wastes from the fuel fabrication plant

Up to now only contaminated trash, packaged in 200 L drums, has been generated. At present there are only four drums in the interim storage area of the facility.

TABLE III. WASTES GENERATED BY ANGRA I

Year	Number of drums containing:					Total number of drums
	Filters	Evaporator concentrates	Non-compressed wastes	Spent resins	Compressed wastes	
1982	14	41	—	—	74	129
1983	17	14	6	—	272	309
1984	8	—	26	73	135	242
1985	10	23	32	60	116	241
1986	22	52	63	2	341	480
1987	11	129	111	—	138	389

4. WASTE MANAGEMENT AT THE IPEN

4.1. Historical development

As a consequence of the small amount of wastes generated until recently in Brazil, the function of waste management at the IPEN was performed mostly by the health physics personnel. Since 1983 a department has been in existence with the following functions:

- To process and treat all kinds of waste generated by the IPEN and some smaller producers;
- To develop new processes or implement processes for treatment of all kinds of radwaste produced in Brazil;
- To realize an extensive research, development and demonstration (RD&D) programme in the field of radioactive waste treatment and disposal.

The Radwaste Department has two branches: one responsible for treatment, conditioning, transport and interim storage, and the other responsible for the RD&D programme and devoted to making available techniques for the treatment of all kinds of waste as well as to implementing a quality assurance programme and performing some waste disposal activities.

4.2. Solid waste treatment facilities at the IPEN

The radwastes generated at the IPEN as well as all the wastes received from medical applications, industry and other institutions are treated by compaction, cementation or incineration or by special techniques developed for specific types of waste.

4.2.1. Compaction facility

Before the installation of the compaction facility the amount of wastes was small and had concentrations below the limits indicated by the radiological regulation. The applied treatment technique was delay and decay prior to disposal to the environment. Later on the solid wastes produced at the IPEN or arriving from other institutions, whether combustible or not, were mostly reduced in volume by simply compacting them directly in the waste drums with a press strength of 10 t. The drums are those used commercially by the chemical industry (200 L capacity). Although compaction results in an average volume reduction factor of only 4 it has low operating costs.

Solid radioactive wastes are produced mainly during cleaning and decontamination activities and consist of rags, paper, cellulose, plastics, gloves, clothing, overshoes, etc. Laboratory materials such as cans, polyethylene bags and glass bottles, as well as bulky exhaust air filters contaminated by the activity adhering to dust particles and aerosols, also contribute to the solid waste inventory.

Small parcels of non-compactible long lived wastes are also produced and received for treatment. They include wood pieces, metal scrap, defective components and tools, and debris from dismantling and decontamination operations. These wastes usually are put in 200 L drums and immobilized by pouring cement paste into the voids.

Because of the widely divergent nature and quality of solid wastes, they must be graded correctly at the place of origin to facilitate optimum treatment. This requires not only separation of combustible and non-combustible items but also a clear separation and classification of beta/gamma and alpha contaminated solid wastes.

Which radionuclides are present in the wastes received at the compaction facility depends on the originating laboratory. The main radionuclides present are: ^{3}H, ^{14}C, ^{24}Na, ^{32}P, ^{35}S, ^{42}K, ^{51}Cr, ^{60}Co, ^{82}Br, ^{85}Sr, ^{90}Sr, ^{95}Mo, $^{99}Tc^{m}$, ^{103}Ru, ^{106}Ru, ^{125}I, ^{125}Te, ^{127}Te, ^{129}Te, ^{131}I, ^{134}Cs, ^{140}Ba, ^{192}Ir, ^{198}Au, ^{226}Ra, U_{nat} and Th_{nat}; in addition there are minor quantities of other radionuclides.

The radioactive wastes are collected at the IPEN in 40 L paper bags which are then placed inside polyethylene bags of 0.2 mm thickness. The closed bags are transported weekly to the treatment facility for compaction and afterwards to interim storage. The non-compactible wastes are also collected in polyethylene bags and labelled for special treatment.

All radioactive waste bags are identified at the waste sources. This is done by the health supervisor, who attaches a properly annotated tag to the transfer bag with the following information: (a) dose rate; (b) radioactive isotopes present; (c) estimate of quantity (Bq or Ci); (d) general description of waste; and (e) location of waste producer.

4.2.2. *Exhausted or defective radioactive sources*

Sealed radiation sources are used in a large variety of equipment in industry, research and medicine and will sooner or later become waste. Most of the sealed sources represent a radiological health problem even when the equipment is no longer in use. For some of the sealed sources used in Brazil the licensees are required by contract with the supplier to take the source back later when the source becomes waste. This is what happens, for instance, with the sources sold by the IPEN, ^{60}Co and ^{192}Ir, or with high activity sources for radiotherapy. A wide variety of sealed radioactive sources (listed in Table I) are sent to the IPEN for treatment. Usually, they are immobilized in concrete drums with their original container.

For gaseous or radium sources specially developed packages were designed. As in other developing countries, Brazil has made extensive use of radium sources in medicine for treatment of cancer tumours. Industrial applications have included radium for radiography, certain electronic valves, switches and luminous paints. The use of radium in all of these applications has been reduced greatly with the growing availability of safer and cheaper radioactive materials, although many radium applications still exist. Owing to the long half-life and decay mode, sealed radium sources will never be disposed of as exempted wastes. Special care has to be taken in order that in interim storage they do not present a hazard to man.

A special package was developed for interim storage, transportation and final disposal of radium sources which can be used for gaseous sources. This special package conforms to the Type A specifications of the IAEA Regulations for the Safe Transport of Radioactive Material [1]. As the final disposal site for LLW and ILW has not yet been defined, the adopted criterion for the package design was that the source should be kept in a form that would not be readily dispersible and that the package could be stored temporarily before transport to the final repository without any hazard to the operating personnel of the interim storage. The package was designed in such a way as to accommodate radium sources of up to 20 GBq ($\sim$0.5 Ci) with a leakage rate lower than 5×10^{-10} kPa·L·s^{-1} to ensure radiological safety.

4.2.3. *Incineration facility for solid wastes*

A system for incinerating combustible solid wastes has been developed in order to achieve higher mass and volume reduction of the wastes generated at the IPEN or received from other institutions. The primary aim of this facility, however, is to burn carcasses of animals used in quality control of radiopharmaceuticals production. This facility, with a designed capacity of 5 kg/h, includes two combustion chambers: an incinerator chamber and an afterburning chamber to ensure complete combustion of gases. The electrical heating adopted provides additional operational safety compared with a gas or oil fired furnace, yielding a further reduction in the volume of off-gases to be treated. The off-gas system utilizes dry treatment and consists of one cyclone, an electrostatic precipitator, a condenser, and an activated carbon filter followed by one HEPA filter. Inactive tests using animal carcasses and before the off-gas system was complete resulted in a burning rate of 2.7 kg/h.

The facility is not yet in operation owing to the need for some major changes in the off-gas system to eliminate flaws. It is expected that it can be put into operation by the end of 1988.

4.3. Liquid waste treatment facilities at the IPEN

A limited volume of liquid wastes containing small quantities of radionuclides is produced at the IPEN as a result of research or radioisotope production activities. This reduced volume results from the optimization of processes to keep the wastes generated to a minimum. Moreover, since most of the radioisotopes handled at the IPEN are short lived the delay and decay technique of treatment is still valid.

Usually the liquid wastes which are not allowed to be discharged directly to the environment are collected in containers and stored for an appropriate time so that the activity decays to an acceptable level. In special circumstances collecting vessels with a maximum capacity of 10 m^3 are constructed in order to permit larger volumes to be managed. Before discharge to the environment some chemical adjustment of the effluent is made; this generates small volumes of sludge but reduces the activity even further below the authorized discharge limits.

4.3.1. *Evaporator–crystallizer unit*

Owing to the characteristics of the iodine production process at the IPEN around 300 L of liquid wastes contaminated with long lived tellurium isotopes are generated annually. Up to now the wastes have been collected in polyethylene bottles and these placed inside steel containers for a decay period of about five years. Evaporation without previous treatment or immobilization was not considered owing to the presence of ^{131}I volatile and high sulphate concentration. The option followed was waste alkalization to pH11 to prevent ^{131}I volatilization and equipment corrosion.

Afterwards an evaporation process is applied for water removal to achieve salt crystallization; this is followed by decay storage in sealed containers. With this procedure the wastes produced in one year can be safely contained in 14 drums.

4.3.2. *Volume reduction of scintillation vials*

Around 3000 L of liquid scintillation vials, used in diagnostic applications and mainly contaminated with ^{3}H, ^{14}C or ^{125}I, are received annually at the IPEN. The total activity is usually lower than 74 MBq (2 mCi).

Because of the small radiological hazard, the current method of treatment, for small quantities, is dilution and dispersion, with care being taken that chemical and radiological discharge limits to the environment are observed. When great volumes are involved the most recommended technique is incineration, if this is available. Special tricks have to be used, however, in order that the long lived radionuclides be trapped in the filters or high dilution be achieved.

A distillation technique is normally used in order to recover solvents (toluene). Quality control procedures show that the recovered solvent is of analytical grade and has no activity, permitting reuse for the same purpose.

4.4. Research and development of techniques for waste treatment and characterization

4.4.1. *Cemented waste characterization*

As soon as activities started in the field of radioactive waste treatment and disposal, some effort was devoted to establishing criteria to control the quality of waste forms.

Cementation was considered a quite reliable process which should be considered for waste immobilization of several types of waste stream in Brazil. Some of the parameters considered for quality control of the immobilized wastes were: homogeneity, solids content, compressive strength, setting time, leach rate, porosity, radiation damage and thermal conductivity. Also considered was the addition of special additives to improve the degree of fixation and the mechanical properties of the solidified products.

One of the most fundamental physical properties required for any kind of immobilized waste form is homogeneity, which is important for the solidification process and long term storage, so it is necessary to specify and define chemical and physical properties such as density, porosity, leach rate, compressive strength, thermal conductivity and radiation damage, which cannot be studied if the matrix is not homogeneous.

Thus, in order to evaluate the degree of homogeneity of simulated wastes immobilized in cement matrices, produced by using a planetary paddle mixer, the

TABLE IV. HOMOGENEITY STATISTICAL TESTS FOR HYDRATED CEMENT BLOCKS[a] WITH DIFFERENT WATER/CEMENT RATIOS AND $NaNO_3$ CONCENTRATIONS IN WHICH URANIUM TRACER WAS USED

Number of hydrated cement blocks	Water/cement ratio	Solids content (wt%)	Average ± standard deviation ($x \pm \sigma$) (ppm)	χ^2 test (A) χ^2	χ^2 test (B) $\chi^2(95\%)$	Kolmogorov test (A) $N^{1/2}D_N$	Kolmogorov test (B) $N^{1/2}D_N(95\%)$	Von Mises test (A) Nw^2	Von Mises test (B) $Nw^2(95\%)$
2	0.30	3.6	106.73 ± 5.96	0.332	5.991	0.52	1.36	0.041	0.461
3	0.30	7.5	124.32 ± 9.42	6.322	7.815	0.52	1.36	0.055	0.461
5	0.35	3.6	101.55 ± 6.47	1.730	5.991	0.83	1.36	0.139	0.461
6	0.35	7.5	95.62 ± 4.91	2.121	7.815	0.56	1.36	0.035	0.461
8	0.40	3.6	99.01 ± 6.51	0.270	5.991	0.51	1.36	0.020	0.461
9	0.40	7.5	110.15 ± 6.14	1.274	5.991	0.64	1.36	0.065	0.461

[a] Number of concentration analyses for each block: 100.
(A) Experimental result.
(B) Theoretical limit.

highly sensitive delayed neutron detection technique was used to measure the distribution of very small quantities of soluble uranium salt or insoluble thorium oxide in powder form [2]. With this procedure the uranium and thorium distributions in the waste form were measured and confirmed to be homogeneous after appropriate statistical tests. These tests are the same as those used to test the distribution of pseudo-random numbers generated by special computer algorithms. The χ^2, Kolmogorov and Von Mises tests were applied, and some results are shown in Table IV.

After the homogeneity of the waste form has been confirmed other properties are analysed to ensure compliance with a set of specified waste form characteristics. The other characteristics measured for simulated nitric wastes immobilized in cement with different solids contents and water/cement ratios were: setting point, compressive strength, hydration temperature, porosity and leach rate of radionuclides.

A study is being carried out to establish a correlation between radionuclide leaching from small scale and large scale specimens. Another study is also in progress where a correlation between porosity and leach rate is proposed that takes into account the actual sample geometry.

In all these studies some effort has been made to evaluate the influence of variations in cement composition; therefore, the chemical composition is always measured.

4.4.2. *Organic solvents*

With the purpose of gaining competence in the treatment of organic solvents remaining after purification of uranium or solvent extraction processes, a well known technique using the incorporation of the solvents in plastics is being studied and tested.

4.4.3. *Final waste disposal*

Some studies are being performed at the IPEN to establish criteria for the land disposal of radioactive wastes. Although some preliminary parameters have already been settled, the final criteria will be available only when the site has been established. Laboratory measurements of radionuclide sorption and migration by batch and column techniques are in progress in order to support the studies for site selection. Long term integrity studies of concrete packages for final disposal are also in progress. More details concerning the final disposal of radioactive wastes in Brazil are presented in another paper at this symposium [3].

REFERENCES

[1] INTERNATIONAL ATOMIC ENERGY AGENCY, Regulations for the Safe Transport of Radioactive Material, 1985 Edition, Safety Series No. 6, IAEA, Vienna (1985).

[2] RZYSKI, B.M., SUAREZ, A.A., Nucl. Chem. Waste Manage. (in press).

[3] FRANZEN, H.R., EIDELMAN, F., PONTEDEIRO, E., IAEA-SM-303/143, these Proceedings, Vol. 1.