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TREATMENT AND CONDITIONING OF INSTITUTIONAL WASTE IN BRAZIL*

Achilles Alfonso Suarez and José de Júlio Rozental[1]

ABSTRACT

An important condition for the expansion of electricity generation from nuclear reactors in the next future is the control of the nuclear fuel cycle. A quite important branch of this cycle is played by the management of radioactive waste. In Brazil there are one nuclear power plant in operation (626 MWe); mining and milling industry, a fuel fabrication plant; three nuclear research institutes including a radioisotope production facility; some accelerators and about 2000 institutions (medicine, industry, agriculture, research centers etc) widespread throughout the country utilizing radionuclides on a large scale. In Brazil there is not a defined final repository to receive the radioactive waste produced in the country. Instead, all radioactive wastes produced must be treated in such a way that:

- the volume of the waste is minimized as much as possible
- the release of radionuclides into the environment is as low as possible
- the radioactive materials are immobilized for storage over long periods safely and preventing them to contact the biocycle.

Discussions are made concerning the brazilian policy and the waste treatment research program carried out by the Instituto de Pesquisas Energéticas e Nucleares (IPEN), scientific branch of the National Commission of Nuclear Energy (CNEN).

TRATAMENTO E CONDICIONAMENTO DE REJEITOS INSTITUCIONAIS NO BRASIL

RESUMO

Uma condição importante para a expansão da geração de energia elétrica produzida por reatores nucleares no futuro próximo é o controle do ciclo de combustível nuclear. Um ramo muito importante deste ciclo é desempenhado pelo gerenciamento de rejeitos radioativos. No Brasil existe uma instalação de energia nuclear em operação (626 MWe); industria de mineração e beneficiamento; uma fábrica de elementos combustíveis; alguns aceleradores e cerca de 2000 instituições (medicina, industria, agricultura, centros de pesquisa etc) espalhados pelo país utilizando radionuclídeos em grande escala. No Brasil não existe ainda um repositório final para receber os rejeitos radioativos produzidos no país. Assim, todos os rejeitos radioativos produzidos devem ser tratados de forma que:

- o volume de rejeitos é minimizado tanto quanto possível
- a liberação de radionuclídeos no ambiente é tão baixa quanto possível
- os materiais radioativos são imobilizados para armazenagem segura por períodos longos de maneira a previnir seu contato com o meio biológico. Dis

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(1) Comissão Nacional de Energia Nuclear-CNEN/Rio de Janeiro, Brasil.

cussões são feitas sobre a legislação brasileira e o programa de tratamento e pesquisa de rejeitos radioativos realizado pelo Instituto de Pesquisas Energéticas e Nucleares (IPEN) associado a Comissão Nacional de Energia Nuclear (CNEN).

1. INTRODUCTION

Production of radioisotopes for medical and industrial use as well experiments in reactor physics, nuclear physics etc for personal training were iniciated in Brazil in the mid-fifties using the experimental reactor IEAR-1-5MW located in São Paulo.

Small amounts of wastes were produced since them which were kept under radiological control till recently. With the installation of the brazilian nuclear power program and increase in the use of radioisotopes in industry, medicine and other fields and with interest in protect public health and ensure safety it was implemented a brazilian regulation covering all aspects of use of radioactive materials either from the nuclear fuel cycle as well from nuclear activities resulting from non nuclear fuel cycle.

For wastes coming from the nuclear fuel cycle great emphasis is given for volume reduction while for those from industry, medicine and research outside the nuclear fuel cycle the volume reduction is no critical because the volumes generated are so small that they do not justify the investment in a volume reduction facility except when they are sent for treatment in institutions which possess such facilities.

In Brazil the main sources of radwastes belong to the category of low level wastes (LLW) and only in few cases medium level wastes (MLW). They are mainly originated from: one nuclear power plant in operation (626 MWe); mining and milling industry; fuel fabrication plant and about 2000 establishments, such as hospitals, industries, research centers widespread throughout the country.

As until the moment there is not a permanent site to receive the radioactive waste produced in the country the LLW and MLW must be treated and stored, in appropriated ways, on the supervision of CNEN, by the producers or sent to IPEN which is in charge of receive those wastes when the radioisotope utilizer does not have ways to do it.

Discussion is made about the brazilian policy and the research and development programme carried out by the IPEN as well the next future concerning final disposal.

2. CLASSIFICATION OF WASTES

The official classification of wastes in Brazil is originated from the IAEA Standards and is entitled "Gerência de Rejeitos Radioativos em Instalações Radioativas - CNEN-NE-6.05-Nov/1985". The classification is based on health and safety requirements according to practical experiences at waste treatment plants from 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, radiation nature, concentration and exposure rate.

Primarily the wastes are classified on the basis of their physical state of aggregation, i.e.

- liquid waste
- solid waste
- gaseous waste

Another very important distinction is the presence or absence of alpha emitters in the waste stream. According to this classification a more appropriate treatment method can be assigned to the waste. In addition to those classifications there is another more detailed classification on the basis of radiological properties of wastes.

For liquid and gaseous wastes this classification is characterized by their 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 are presented in Table I.

The elimination of certain amounts of wastes from any installation is conditioned to particular authorization to be obtained from the national Commission of Nuclear Energy. The upper limit for the specific activity of solid wastes permited to be disposed in the urban collection system is 74 Bq/g (2 nCi/g) while for liquid and gaseous wastes allowed to be eliminated in sewage or atmosphere a specific table containing those informations for each radionuclide is provided in the regulation.

According to this regulation persons engaged in activities utilizing radioactive materials must provide complete informations and technical data on training, personnel experience, safety procedures, operation system of interim storage, disposal of efluentes and detailed analysis of this proposed operation.

The applicant to use of radioisotopes may request to CNEN for specific approval of proposed procedure to dispose of licensed radioactive material in a manner other than as generaly authorized in the regulations. If necessary, meetings are held between the CNEN staff and the applicant staff in order to solve specific questions.

To assure compliance with the approved conditions all licenses are subject to periodic inspection. Radioactive waste, which is not more possible to be under responsability of the applicant are collected by the CNEN inspectors and sent to IPEN for the 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 medical and biochemical research fields as well as in the clinical area, together with the wastes resulting from the application of radioactive material in industrial processes are treated as follow:

- for short half-lived radionuclides such ^{125}I, ^{131}I, ^{67}Ga, ^{201}Tl, ^{32}P, ^{35}S, ^{99m}Tc, the majority originated from medical and research institutions is employed a decay treatment method followed of disposal by release to the sanitary sewage.
- 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 excretion (faeces, urine, vomite) of patients who have received therapeutic doses in excess of 555 MBq (15 mCi) has to be collected in bags and, after decay, emptied into a sink for excreta.
- sources of radiotherapy must be returned to the country of origin as well the sources used in sterilization.
- those sources generally used in industrial equipments as for foil thickness measurements, level detection, gauging, quality control, smoke de-

tectors etc are generally sent to IPEN for treatment.

- exausted ^{192}Ir sources mostly used in non-destructive testing are sent also to 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, sealed sources and targets. Miscellaneous liquids containing short-lived isotopes or small concentrations are generally released to the sewer. Trash, sealed sources and targets are normally packaged in 100 or 200 L drums. IPEN give special attention to the users of ^{3}H and ^{14}C contained in liquid scintillation vials and some animal carcasses, in compliance with the regulations.

3.3. Wastes from the Brazilian Nuclear Power Plant Angra I

The operation of nuclear power plants does not produce excessive amounts of waste if one considers the total fuel cycle; most of the waste produced by them fall exclusively under 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 II indicates approximately the volumes of radioactive wastes and activities generated by the nuclear power plant of Angra I in operation.

3.4. Wastes from the fuel fabrication plant

Until now only contaminated trash, packaged in 200 L drums were generated. At moment there are only four drums at the interim storage area in the facility.

4. WASTE MANAGEMENT AT NUCLEAR AND ENERGY RESEARCH INSTITUTE (IPEN)

4.1. Historical development.

As a consequence of the small amount of waste generate till recently in Brazil, the function of waste management at IPEN was performed mostly by the health physics personnel. After 1983 a Department was established with the following functions:

- to process and treat all kind of waste generated by IPEN and from some smaller producers

- to develop new processes or implement processes for treatment of all kinds of radwastes produced in Brazil .
- to realize an extensive research, development and demonstration program, RD&D, in the field of radioactive waste treatment and disposal.

That department has two branchs: one responsible for the treatment, conditioning, transport and interim storage and the other responsible for the RD&D program devoted to make available the techniques for the treatment of all kinds of waste as well to implement a quality assurance program and realize some activities in the field of waste disposal.

4.2. Treatment facilities for solid waste at IPEN

The radwaste generated at the institute as well all the waste received from medical applications, industry or other institutes are treated at IPEN using either compaction, cementation, incineration or any other technique specially developed for the specific waste.

4.2.1. Compaction unit

Before the installation of the compaction facility the amount of waste material was small and had concentrations below the limits indicated by the federal radiological regulations. The applied treatment technique was delay and decay prior dispose to the environment. Later on the solid waste produced at IPEN or arriving from other institutions, whether combustible or not, is mostly reduced in volume by simple compacting directly in the waste drums with a press strength of 10 ton. The drums are those usually commertialized by the chemical industry (200 L capacity). Although compaction results in average volume reduction factors of only 4:1 it is characterized by low operating costs.

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

A small parcel of non compactible long lived wastes are also produced and received for treatment. They are wood pieces, metal scraps, defective components and tools, debris from dismantling and decontamination oper ations etc. 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 waste, it must be graded right at the place of origin to allow optimum methods of treatment to be applied. These requirements extends no only to the different nature of the waste in terms of burnable and non-burnable, but also to a clear separation into and classification as β-γ contaminated and α contaminated solid wastes.

The radionuclides present in the wastes received at the compaction facility of IPEN depends from which laboratory they arrive. The main radionuclides present in the wastes 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, ^{99m}Tc, ^{103}Ru, ^{106}Ru, ^{125}I, ^{125}Te, ^{127}Te, ^{129}Te, ^{131}I, ^{134}Cs, ^{140}Ba, ^{198}Au, ^{226}Ra, U-nat, Th-nat and minor quantities of other radionuclides.

In Fig. 1 is shown the relative contribution of the main solid waste contaminants and in Fig. 2 is shown the relative contribution according to the waste source.

The radioactive wastes are collected in 40 L paper bags placed inside to polyethylene bags of 0,2 mm thickness. The closed bags are transported weekly to the treatment facility for compaction and after to interim storage. The non-compactible wastes are collected also in polyethylene bags and addressed for special treatment.

All radioactive wastes bags are identified at the waste sources. This is done by the health superviser which attaches a properly filled tag to the transfer bag with the following information: a) level of activity; 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 equipments used in industry, research and medicine and will sooner or later become waste. Most of the sealed sources do represent a real radiological health problem even when the equipment is no longer in use.

For some of the sealed sources used in Brazil there is a contract with the supplier of the source to take it back at a later time when the source becomes waste. This is what happens for instance with the sources sold by IPEN (^{60}Co, ^{192}Ir).

A relatively long list of sealed radioactive sources shown in Table III, is usually sent to IPEN for treatment. Usually they are immobilized in concreted drums with their original container.

For those 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 by insertion of encapsulated needle sources directly into the tumor or by means of moulded applicators that held the source next to the tumor. 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 availability of safer and cheaper radioactive materials although many radium applications still exist.

Due to its long half life and decay mode sealed radium sources shall never be disposed as exempted waste. Even more special care has to be taken in order that in interim storage they do not represent an hazard to man.

A special package was developed for interim storage, transportation and final disposal of radium sources which can be successfuly used for gaseous sources. This special package conform to type A - non special form, of the Regulations for the safe transport of radioactive materials(1). As the final disposal site has not yet been defined, the adopted criterion for the package design was to keep the source in a non readily dispersible form which could be kept temporally stored 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 to accomodate radium sources with 20 GBq (≈0,5 Ci) with a leakage rate lower than 5×10^{-9} mbar.L/s assuring 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 IPEN or received from other institutions. The primary aim of this facility however is to burn animal carcasses resulting from quality control of radiopharmaceuticals production. This facility with a designed capacity of 5 Kg/h includes two combustion chambers: incinerator chamber and after-burning chamber in order to assure complete combustion of gases. The electric heating adopted provides additional operational safety compared to gas or oil fired furnace, resulting further in the reduction of volume of off-gases to be treated. The off-gas system utilizes dry treatment and consists of one cyclone, electrostatic precipitator, condenser, activated carbon filter followed by one HEPA filter. Inactive tests using animal carcasses and prior the off-gas system be complete resulted in a burning rate of 2.7 Kg/h.

The facility is not yet in operation due to some drastic changes in the off-gas system intended to eliminate some flaws. It is expected that it can be operating by the end of 1988.

4.3. Treatment facilities for liquid wastes at IPEN

Limited volumes of liquids wastes containing small quantities of radio nuclides are produced at IPEN as a result of research or radioisotope production activities. This reduced volume of liquid wastes results from the optimization of processes in order to keep to a minimum the wastes generated. Even more, since most of radioisotopes handled at 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 for a period of time that allow the present radionuclide to decay to an acceptable level. In special circumstances collecting vessels of 10 m^3 capacity in maximum are constructed in order to permit larger volumes to be managed. Before the discharge to the environment some chemical adjustment of the effluent is made generating small volumes of sludge but reducing even more the activity bellow the authorized discharge limits.

4.3.1. Evaporator/crystallizer unit

Due to the characteristics of the iodine production process used at IPEN around 300 L of liquid waste contaminated with long-lived telurium isotopes are generated. Until now those wastes are collected in polyethylene bottles placed inside steel containers to decay for about 5 years. The evaporation without previous treatment or immobilization were not considered due to the presence of ^{131}I volatile and high sulfate concentration. The option followed was waste alkalinization until pH = 11 to prevent ^{131}I volatilization and equipment corrosion. Afterwards an evaporation process was applied for water reduction until salt crystallization followed by storage in sealed containers for decay. With this procedure the waste produced in one year can be safely contained in some drums.

4.3.2. Volume reduction of scintillation cocktails

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

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

A reflow distillation technique was used instead aiming to reduce the volume of waste to be treated as well recovering the solvent. The elimination of the solvent to the environment should be precluded since it is a carcinogenic agent. This technique was applied for cocktails using toluene as solvent.

Quality control procedures shows that the recovered solvent is analytical graded and has no activity in it permiting to be reused again 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 has been devoted to establish criteria to control the quality of waste forms.

Cementation was considered a quite reliable process which should be considered for waste immobilization of several waste streams in Brasil. Some of the parameters considered for quality control of the immobilized waste were: homogeneity, salt contents, compressive strength, setting time, leaching rate, porosity, radiation damage, thermal conductivity etc. It was considered also 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 property required for any kind of immobilized waste form is the homogeneity which is quite important in the solidification process and during long-term storage. It is the starting point to specify and define some chemical and physical properties as density, porosity, leaching rate, compressive strength, thermal conductivity, radiation damage etc which can not be studied if the matrix is not homogeneous.

Thus, in order to evaluate the degree of homogeneity of simulated wastes immobilized in cement matrices, by using a planetary paddle mixer, it was used the high sensibility of the delayed neutrons detection technique to measure the distribution of very small quantities of soluble uranium salt or insoluble thorium oxide, in powder form[(2)]. With this procedure it was measured the uranium and thorium distribution along the waste form which was confirmed to be homogeneous, by the application of appropriated statistical tests. Those statistical tests are the same used to test the uniformity of pseudo-random numbers generated by special algorithms used in computers. For that purpose it was applied more than one statistical test, namely the χ^2, Kolmogorov and Smirnov-Cramer-Von Mises tests. Some results obtained by using such technique is shown in Table IV.

After the homogeneity of the waste form be confirmed other properties are analysed aiming to assure compliance with a set of specified waste form characteristics. The other characteristics measured for simulated nitric wastes immobilized in cement according the salt contents and water to cement ratio were: setting point, compressive strength, hydration temperature, porosity, leaching of radionuclides etc.

Particularly in terms of leaching a study is been realized aiming to establish a correlation between radionuclide leaching from small scale and large scale specimens. Other study is also in progress where a correlation between porosity and leaching rate is proposed taking into account the actual sample geometry.

In all these studies some effort has been made to evaluate the influence of the variations from cement composition. With this purpose in mind details of the cement chemical composition is always measured.

4.4.2. Organic solvents

With the purpose of to get competence for treatment of organic solvents resulting from purification of uranium or solvent extraction processes a well known technique using the incorporation in plastics is been studied and tested.

4.4.3. Final waste disposal

Some studies are been realized at IPEN in cooperation with CNEN to establish criteria for the land disposal of radioactive waste. In spite of some preliminary parameters be already settled the final criteria will be available only when the site be established. Laboratory measurements of radionuclide sorption and migration by using 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.

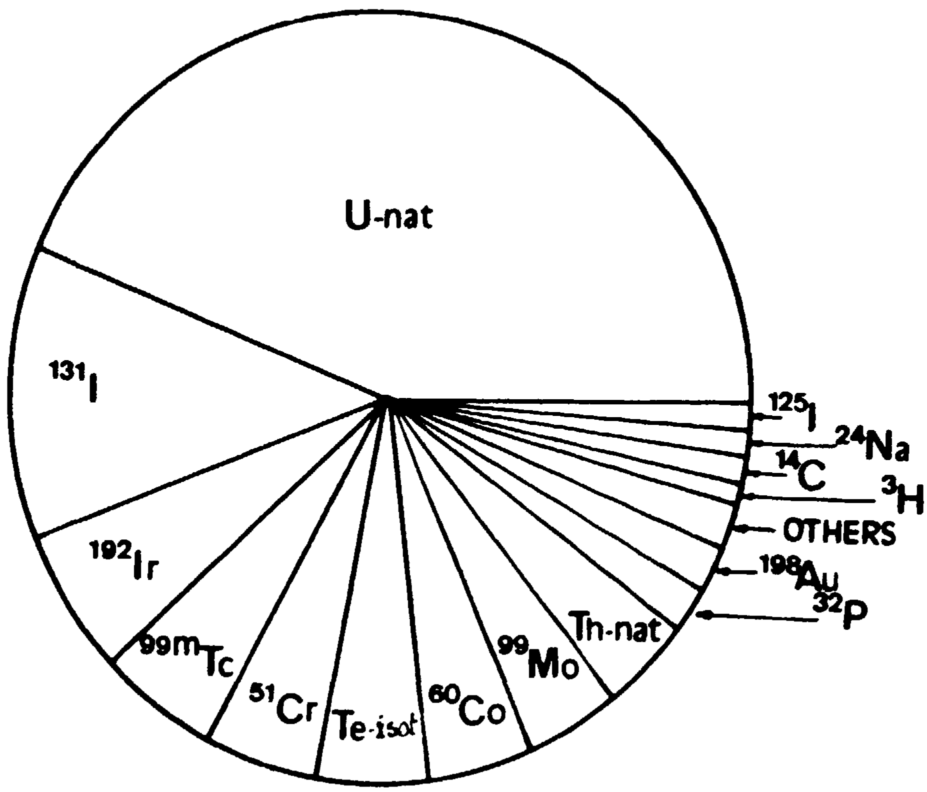

Figure 1 – Solid waste contaminants, volume %

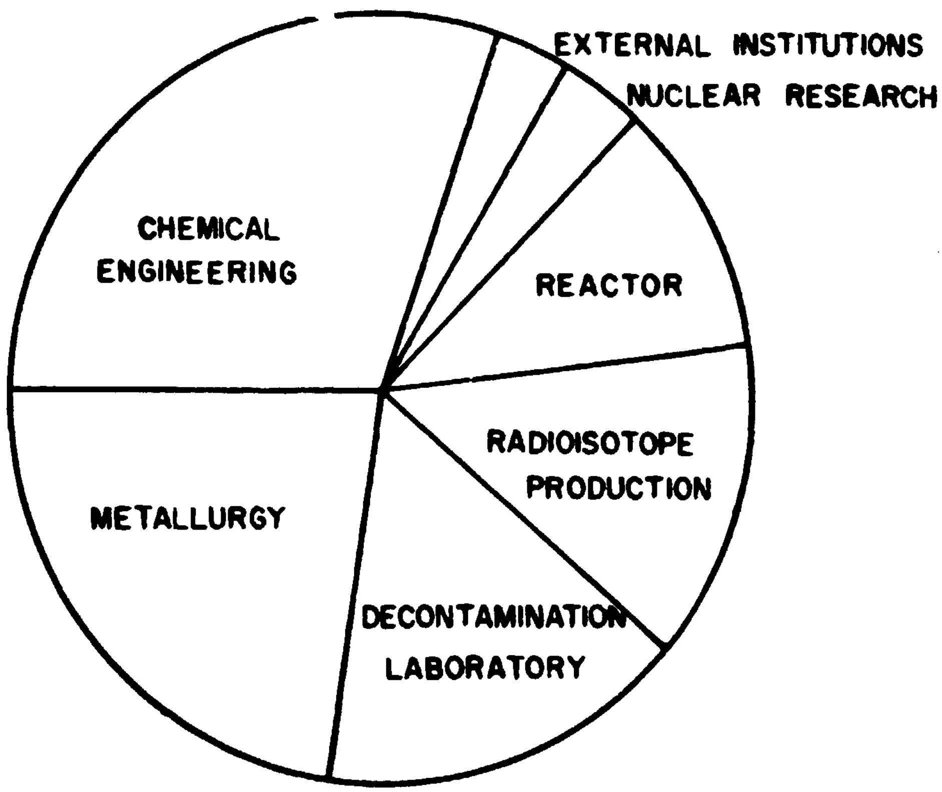

Figure 2 – Waste producers, volume %

TABLE I - CLASSIFICATION OF RADIOACTIVE WASTES IN BRAZIL

Category	Beta, gamma emitters (α emission < 3.7×10^{8} Bq/m^3)	Alfa emitters (α emission > 3.7×10^{8} Bq/m^3)
	Surface exposure rate ($\dot{X}$) (μC/kg.h)	Concentration (c) (Bq/m^3)
I. Solid wastes		
Low level	$\dot{X} < 50$	$3.7\times10^{8} < c < 3.7\times10^{11}$
Medium level	$50 < \dot{X} < 500$	$3.7\times10^{11} < c < 3.7\times10^{13}$
High level	$\dot{X} > 500$	$c > 3.7\times10^{13}$
	Concentration (c) (Bq/m^3)	Concentration (c) (Bq/m^3)
II. Liquid wastes		
Low level	$c < 3.7\times10^{10}$	$3.7\times10^{8} < c < 3.7\times10^{10}$
Medium level	$3.7\times10^{10} < c < 3.7\times10^{13}$	$3.7\times10^{10} < c < 3.7\times10^{13}$
High level	$c > 3.7\times10^{13}$	$c > 3.7\times10^{13}$
	Concentration (c) (Bq/m^3)	
III. Gaseous wastes		
Low level	$c < 3.7$	
Medium level	$3.7 < c < 3.7\times10^{4}$	
High level	$c > 3.7\times10^{4}$	

TABLE II - WASTES GENERATED IN THE POWER PLANT ANGRA-I

Year	Number of drums with					Total amount of drums
	Filters	Evaporator concentrates	Non compressed wastes	Spent resins	Compressed wastes	
1982	14	41	-	-	74	129
1983	17	14	06	-	272	309
1984	08	-	26	73	135	242
1985	10	23	32	60	116	241
1986	22	52	63	02	341	480
1987	11	129	111	-	138	389

TABLE III - SEALED SOURCES USED IN INDUSTRY, RESEARCH AND MEDICINE RECEIVED BY IPEN

Source	Source strength (G Bq)	Half life (years)	Principal applications
^{3}H	1 - 1000	12.3	Production of neutrons by (D, T) reactions
^{60}Co	up to 4 x 10^{3}	5.3	Radiotherapy, industrial radiography, level gauge
^{85}Kr	0.1 - 50	10.8	Paper thickness measurements
^{90}Sr	0.1 - 2	29.1	Paper thickness measurements, beta therapy
^{137}Cs	up to 4 x 10^{3}	30.1	Radiotherapy, industrial radiography, density level gauge
^{147}Pm		2.6	Paper thickness measurements
^{241}Am	1 - 10	432	Density gauge, lightning-rod
^{241}Am-Be	0.1 - 10	432	Moisture detector

TABLE IV - HOMOGENEITY STATISTIC TESTS FOR HYDRATED CEMENT BLOCKS[a], WITH DIFFERENT W/C AND $NaNO_3$ CONCENTRATIONS, WHERE URANIUM TRACER WAS USED

Hydrated cement block	Water/cement	Salt content (wt%)	Average ± standard deviation ($\bar{x} \pm \sigma$) (ppm)	χ^2 test: χ^2 (A)	χ^2 test: χ^2(95%) (B)	Kolmogorov test: $\sqrt{N}\, D_N$ (A)	Kolmogorov test: $\sqrt{N}\, D_N$(95%) (B)	Von Mises test: Nw^2 (A)	Von Mises test: Nw(95%) (B)
2	0.30	3.6	106.7 ± 6.0	0.33	5.99	0.52	1.36	0.04	0.46
3	0.30	7.5	124.3 ± 9.4	6.32	7.82	0.52	1.36	0.06	0.46
5	0.35	3.6	101.6 ± 6.5	1.73	5.99	0.83	1.36	0.14	0.46
6	0.35	7.5	95.6 ± 4.9	2.12	7.82	0.56	1.36	0.04	0.46
8	0.40	3.6	99.0 ± 6.5	0.27	5.99	0.51	1.36	0.02	0.46
9	0.40	7.5	110.2 ± 6.1	1.27	5.99	0.64	1.36	0.07	0.46

(A) experimental

(B) theoretical limit

[a] number of concentration analysis for each block = 50

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