REUTILIZATION AND CONDITIONING OF SPENT SEALED SOURCES FROM NUCLEAR GAUGES

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ABSTRACT

The increase in use of nuclear gauges in Brazil has resulted in a corresponding increase in the volume of radioactive waste, that must be stored and conditioned for disposal. After being taken out of service, the spent sources are classified as waste and sent to research institutes that are part of the National Commission of Nuclear Energy - CNEN, which are responsible for receiving, storing and conditioning such waste for disposal. CNEN has been intensifying efforts to collect such spent nuclear gauges from the users; in order to minimize the possibility of accidents. As a result of these efforts, a large number of sources are stored at the CNEN institutes.

The management goal for these sources is minimize the waste that must be stored and disposed, through reutilization or a more efficient method of disposal conditioning. The guideline being used is reutilize those sources in which the radioactive material exists as special form radioactive material and that do not exhibit leakage. Nuclear gauges whose sources do not meet these requirements will be dismantled and the sources efficiently conditioned. To dismantle the devices a hot-cell is being implemented at the Centro de Desenvolvimento da Tecnologia Nuclear - CDTN. The hot-cell will allow dismantling of ^{60}Co nuclear gauges with activities up to 7,4 GBq (200 mCi) and ^{137}Cs up to 740 GBq (20 Ci).

This management strategy is expected to reduce the number of 200-L drums of conditioned waste from 92 to 11, when compared with conventional methods of conditioning.

INTRODUCTION

There are a number of different definitions for the term "sealed radiation source". In the IAEA Glossary [1] a sealed source is defined as:

"A source whose structure is such as to prevent, under normal conditions of use, any dispersion of the radioactive material into the environment." A spent radiation source is then a source for which no further use is foreseen.

Many applications of radiation sources require the activity to be concentrated into as small a volume as possible, usually on order of a cubic centimeter or a few cubic millimeters, which gives the source very small dimensions even though the overall volume is increased by encapsulation [2]. In modern practice the radioactive material inside the encapsulation should be in an insoluble form, for example metal in the case of ^{60}Co and ^{192}Ir or a ceramic for ^{137}Cs and ^{241}Am. The material used for encapsulation is usually stainless steel, but sometimes platinum, titanium, or other metals are used. Commercially produced sources are manufactured and tested to internationally agreed upon standards [3,4].

The utilization of nuclear gauges in Brazil has increased in recent years mainly because of the following factors:

- competitive cost when compared with conventional gauges;
- growing presence of multinational companies that are used to using nuclear measurements techniques;
- easy control and measurement of the desired variables such as level, thickness, density, etc;
- absence of contact between the material whose level or density requires measurement and the gauge, avoiding problems of corrosion and abrasion;
- availability and use of high sensitivity detectors, making possible the use of smaller sources.

After being taken out of service, the sources are classified as radioactive waste by their users and sent to the CNEN research institutes, that are responsible to receive, store and, when necessary, condition the waste. Normally, a source is removed from service when its activity has decreased to a value below a preset minimum, when the equipment or the technique used become obsolete or when the equipment is damaged.

Accidents with spent radiation sources have been more severe than those involving sources still in service, because the latter are discovered quickly in opposition to the former. The accidents generally occur due to inadequate handling by unqualified persons, that do not know anything about radioactivity and its effects. Because of this, CNEN has been intensifying its source collection efforts in the sense of picking up spent sources from the users; in order to minimize the possibility of accidents [5].

The management strategy for these sources consists of waste minimization, through the reutilization of the nuclear gauges or a more efficient methodology for conditioning the spent sources. It is intended to reutilize those sources originally classified as special form radioactive material and that do not exhibit leakage. The devices whose sources do not meet these requirements will be dismantled and the sources conditioned efficiently. To dismantle the devices a hot-cell is being implemented at CDTN. Using the hot-cell, it will possible to dismantle ^{60}Co nuclear gauges with activities up to 7,4 GBq (200 mCi) and ^{137}Cs nuclear gauges with activities up to 740 GBq (20 Ci).

This paper describes the nuclear gauges received at CDTN, located in Belo Horizonte / MG-Brazil. The process that will be used for reutilization of the nuclear gauges and the methodology that will be used for conditioning of the spent sources also are described.

NUCLEAR GAUGES

In Table 1 are briefly described the main applications involving nuclear gauging techniques and typical sources are listed [6].

Table 1. Main Applications Involving Nuclear Gauges

Technique	Principle	Radiation Source	Typical Uses
Transmission Gauges	The source of radiation and the detector are on opposite sides of the sample under investigation. Radiation is transmitted through the sample and the degree of absorption is a direct measure of the thickness or density. The radiation quality and intensity are adjusted to each particular application.	Pm-147 Kr-85 Tl-204 Sr/Y-90 (activities 40 MBq to 40 GBq) Medium voltage X rays Am-241 Cs-137 (activities 0,4-40 GBq)	Thickness measurements of plastic, paper, metal sheet, rubbers, textiles, etc. Determination of tobacco content of cigarettes Measurement of dust and pollutant levels on filter paper samples. Monitoring of bulk flow of material on conveyors or in pipes Bone density measurement to diagnose osteoporosis.
Beta Backscatter Gauge	The source and detector are on the same side of the sample, often with the source incorporated into the face of the detector. The intensity of the radiation backscattered from thin samples gives a measure of the sample thickness and/or the atomic number of the sample.	Pm-147,Kr-85 Tl-204 and Sr-90/Y-90. For point sources the activities usually fall within the range 40-200 MBq.	Measurement of the thickness of thin samples of paper, plastics, rubber, etc. Determination of the thickness of a coating on a substrate, provided that there is sufficient difference in either the densities or the atomic numbers of the two materials.

Gamma Backscatter Gauge	The source and detector are mounted on the same side of the sample and the intensity of the scattered radiation is a measure of the sample thickness or the mean atomic number of the material. The method is used for the measurement of substances of low atomic number for which transmission measurements are not sufficiently sensitive.	Am-241 (4GBq) Cs-137 (2 GBq) Pu-238 (1 GBq)	Thickness determination for light alloys, glass, plastics, rubbers, etc., beyond the range of measurement of beta sources Measurement of the wall thickness of pipes, tanks, process vessels, etc. Measurement of the ash content of coal (the ash increases the mean atomic number) Coal sensing probes on automatic coal cutting machines.
X Ray Fluorescence Gauge	A low energy photon source is used to excite fluorescent X rays in either the substrate or the coating of the material under investigation. The fluorescent X rays from the metal substrate are absorbed as they pass through the plastic coating and their resultant intensity is a measure of the thickness of the coating.	Fe-55 (200-800 MBq) Tritium (40 GBq) Pu-238 (1 GBq) Am-241 (4-40 GBq)	Measurement of the thickness of plastic coatings on metals Measurement of the thickness of metals on other metals where there is only a small difference in atomic number between the metals, e.g. tin and zinc on iron, precious metals on copper.
Photon Switching (Level Gauge)	Gamma rays or X rays are transmitted from one side of a container or vessel to a detector mounted on the opposite side or sometimes from a source within the vessel to a detector on the outside. The intensity of the radiation reaching the detector is dependent upon the level of the contents of the vessel.	Low to medium energy X rays (liquids in cans) Am-241 (4 GBq), Cs-137 and Co-60 (often in the range 2-80 GBq).	Packaging monitors, providing automatic rejection of partially filled cans, bottles, etc. Process control of the contents of large vessels, hoppers, etc., e.g. liquids in vessels, coal in hoppers Alignment of pusher and container on coke ovens.

Selective Gamma Absorption	X-rays or gamma rays of a specific energy are transmitted through the sample under investigation. The energy of the radiation is chosen so that absorption in the sample is predominantly sensitive to one compound or element	Am-241/Ag (400 MBq) Fe-55 (800 MBq)	Measurement of sulphur in oils Measurement of lead in petrol Measurement of ash in paper.
Gamma Scattering	When gamma rays from a high-energy gamma source are backscattered from a medium, their intensity gives a measure of the bulk density and porosity of the medium	Cs-137 or Co-60,at activities of about 20 GBq.	Typical uses include soil density measurements both in boreholes and on the surface.
Thermalization of Neutrons	Fast neutrons interacting with the sample medium are slowed down to epithermal or thermal energies by collisions with hydrogen or other light elements in the sample. The intensity of the slowed down neutrons gives a measure of the concentration of these elements.	Am-241-Be (1-800 GBq) Cf-252 (0.1 μg).	Soil moisture measurements in agriculture and in the construction industry (detection of the hydrogen content of the moisture). Oil prospecting (primarily detection of the hydrogen content of oil deposits). Production control measurement of the moisture content of such materials as wood chips and sintermix coke.
Neutron Transmission	Thermal neutrons are transmitted through the sample under investigation and are removed out of the transmitted beam either by absorption or by scattering. The resultant intensity of the transmitted beam then gives a measure of elements in the sample, which have a high cross-section for either neutron absorption or neutron scattering.	Am-241-Be (10-40 GBq) Cf-252 (approx. 10 μg).	Measurement of boron content of detergents, glass, etc. Bulk element analysis in process control, e.g. water, lithium, boron, cadmium Weighing of explosives.

NUCLEAR GAUGES RECEIVED AT CDTN

A database is being implemented in order to facilitate the organization and retrieval of information about nuclear gauges stored at CDTN. Examples of the information being included are [7] :

- radionuclide, activity and date (e.g. ^{60}Co, 1 GBq, January 14th 1988);
- physical and chemical form of radionuclide;
- producer of the source;
- source type including dimensions and shape;
- source serial number;
- user of the source including name and address of person knowing details of the equipment i.e. dimensions, material, etc.;
- results of tests which have been done, for example leak tests;
- details of the equipment, i.e. dimensions, material, etc.;
- measured dose rates (at the surface and at 1-m distance from it);
- whether the source was originally conditioned as special form radioactive material;
- working life of the equipment, etc.

The reutilization of nuclear gauges and the conditioning of spent sources will be planned based on this data.

Table 2 summarizes the source radioactivity and number of nuclear gauges received at CDTN until April 30th 1998.

Table 2. Nuclear Gauges Received at CDTN as Radioactive Waste

ISOTOPE *	NUMBER OF SOURCES	APPROX.TOTAL ACTIVITY GBq (Ci)
^{241}Am	03	110 (3.0)
^{60}Co	109	40 (1.08)
^{137}Cs	78	3470 (93.8)
^{85}Kr	19	70 (1.9)
^{238}Pu, ^{14}C, ^{3}H, ^{147}Pm, ^{63}Ni, etc.	83	80 (2.2)
Total	292	3770 (102)

* Only isotopes with half-life greater than one year

REUTILIZATION OF NUCLEAR GAUGES

It is intended to reutilize only sealed sources containing certified special form radioactive material. Sources received at CDTN with this certification will be wipe tested in a hot-cell. A special Geiger Müller detector for measuring very low level radiation will be used to detect leakage at detection levels, even lower than the established leakage limit in the CNEN-NE-5.01 Standard

[8]. If the source does not exhibit leakage and the shutter operation and shielding of the nuclear gauge is intact, it will made available for reuse at a cost savings to the user over the purchase of a new source.

To be classified as special form radioactive material, specimens that comprise or simulate the material will be subjected to an impact test, a percussion test, a bending test, and a heat test [9]. A different specimen may be used for each of the tests. Following each test, a leaching assessment or volumetric leakage test should be performed on the specimen.

CONDITIONING OF SPENT SEALED SOURCES

Conditioning is defined by IAEA [10] as "those operations that produce a waste package suitable for handling, transportation, storage and/or disposal. Conditioning may include the conversion of the waste to a solid waste form, enclosure of the waste in containers and, if necessary, providing an overpack".

IAEA recommends direct conditioning of nuclear gauges containing spent sources in concrete [11]. This typically limits the number of spent sources to be incorporated in a drum to two to four, as shown in Fig. 1.

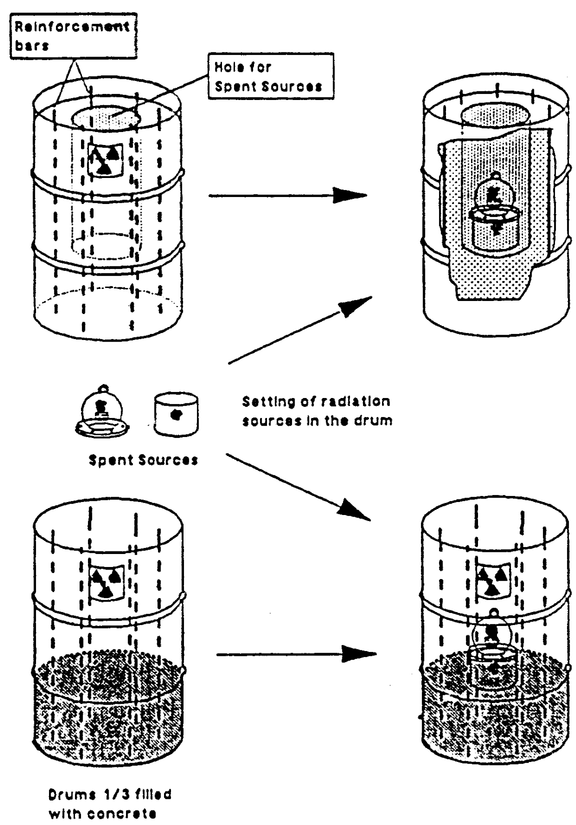

Fig. 1. Conditioning of Spent Sealed Sources in Type A Packages

STANDARDS ASPECTS OF CONDITIONING OF SPENT SEALED SOURCES

In Brazil there are no specific standards related to spent source conditioning. There is a proposed standard "CNEN-NE-6.09 - Acceptance Criteria for Low and Medium Level Radioactive Wastes" [12], that establishes criteria for acceptance of radioactive waste in the future Brazilian Repository. The proposed standard establishes that the final packaging should comply with the National Standard for Transport of Radioactive Material [8] and the Requirements for Safe Disposal.

IAEA recommends that the maximum activity of a package containing beta-gamma isotopes to be disposed by shallow land burial, should be less than 740 GBq (20 Ci) [13].

Transport of dangerous goods is regulated in Brazil by "96044 Decree" of 18/05/88; transport of radioactive materials should also comply with the specific CNEN standard. CNEN radioactivity limits per Type A package are presented in Table 3, where A_1 is the limit of activity if the radionuclide exists as special form radioactive material; and A_2 for other than special form radioactive material [8].

Table 3. Values of A_1 and A_2 for More Used Radionuclides

Radionuclide	A_1 - GBq (Ci)	A_2 - GBq (Ci)
^{137}Cs	1850 (50)	370 (10)
^{60}Co	370 (10)	370 (10)
^{192}Ir	740 (20)	370 (10)
^{241}Am	1850 (50)	0,185 (0,005)
^{226}Ra	296 (8)	18,5 (0,5)

METHODOLOGY FOR CONDITIONING OF SPENT SOURCES AT CDTN

The CDTN guidelines for establishing a methodology for conditioning spent sources are:

- Compliance with the limits of the transport standard;
- Use of Type A packaging;
- Possible retrieval of the sources, if it is necessary;
- Conformance with the maximum activity limit recommended by IAEA for disposal of waste containing beta-gamma radionuclides;
- Low doses of radiation for involved workers;
- Use of a high performance concrete to provide high compressive strength and low permeability [14];
- Implement an immobilization facility if the final safety assessment of disposal, attests to the suitability of the methodology.

The methodology is illustrated in the Figure 2.

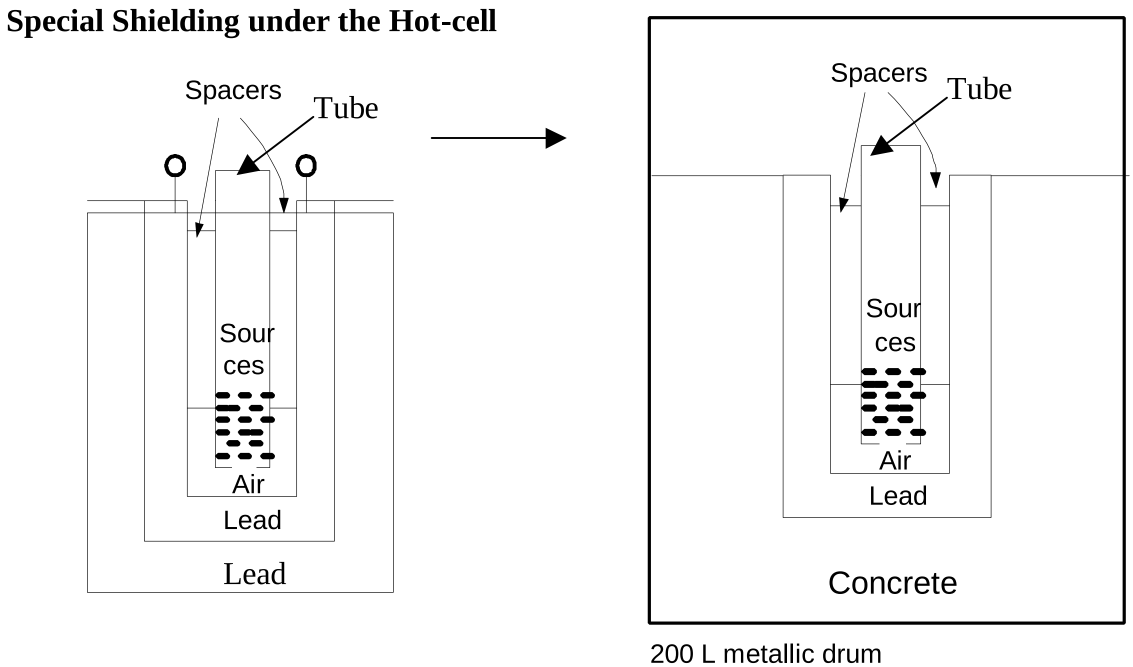

Fig. 2. CDTN Spent Sources Conditioning Process

The methodology, illustrated in Figure 2, includes the following steps:

1. dismantle the nuclear gauges in the hot-cell;
2. transfer of sources to an air, lead-, and concrete-shielded tube below the hot-cell, until the limit of activity is reached;
3. place a top seal on the shielded tube;
4. prepare a drum with internal concrete shielding as shown;
5. transfer the tube with shielding to the drum below the hot-cell;
6. place a concrete cap on the drum, providing shielding and isolation.

At this point the sources will not be immobilized, so as to retrievable if necessary. The final conditioning will only be accomplished after the establishment of the acceptance criteria for radioactive waste in the national repository. If the methodology complies with the criteria, immobilisation would be accomplished as follows:

7. remove the concrete cap;
8. fill the space around the sources with a metallic alloy with low melting point;
9. seal the drum with fresh concrete.

The utilization of metallic alloy in the immobilization process presents the advantage of very low permeability, high mechanical resistance and low corrosiveness since compatible materials are used [15]. In addition, this provides an extra measure of safety for an intrusion scenario.

Various thicknesses of concrete and lead were evaluated using the software "Microshield", for thickness of air fixed at 2.5 cm. A maximum activity of 74 GBq (2 Ci) of ^{60}Co and 370 GBq (20 Ci) of ^{137}Cs was assumed - about 5 and 2.5 times lower, respectively, than the transport standard limits. The radiation levels at the external surface of the drum were calculated for filled heights of the tube cavity being 10, 20 and 30 cm. The range of filling heights was chosen to account for variable geometries and the activities of the individual sources, i.e., high activity sources may cause the package radioactivity limit to be reached before the 30 cm available tube cavity is filled. The results are presented in Tables 4 and 5.

Table 4. Variation of Radiation Level According to Source Filling Height - ^{60}Co

Concrete Thickness (cm)	Lead Thickness (cm)	Estimated Weight of Drum (kg)	Radiation Levels at External Surface of Drum (mSv/h)		
			10 cm Height	20 cm Height	30 cm Height
20.0	3.0	540	**13130**	11960	10460
18.5	4.5	570	**6790**	6120	5280
17.0	6.0	610	**3480**	3100	2640
15.5	7.5	660	**1770**	1560	1310
14.0	**9.0**	**710**	**890**	**780**	**650**

The packaging that will be used for conditioning of ^{60}Co sources will include 14.0- cm -thick-concrete and 9.0 - cm - thick-lead, the most conservative situation (shown in red).

Table 5. Variation of Radiation Level According to Source Filling Height - ^{137}Cs

Concrete Thickness (cm)	Lead Thickness (cm)	Estimated Weight of the Drum (kg)	Radiation Levels on the External Surface of the Drum (mSv/h)		
			10 cm Height	20 cm Height	30 cm Height
22.0	1.0	510	**24940**	22630	19690
21.0	2.0	530	**11010**	9862	8441
20.0	3.0	550	**4797**	4245	3577
19.0	4.0	575	**2070**	1810	1503
18.0	**5.0**	**610**	**887**	**766**	**627**

As in the case of ^{60}Co, the most conservative situation is the use of packaging with 18-cm-thick concrete and 5-cm-thick-lead.

HOT-CELL FOR DISMANTLING NUCLEAR GAUGES

A hot-cell is being implemented in order to dismantle nuclear gauges received at CDTN, allowing removal of the radiation sources from their original shielding.

A hot-cell can be defined as a heavily shielded enclosure for handling and processing (by remote means or automatically) or storing highly radioactive materials. The main function of the hot-cell is to allow safe handling of high radiation sources, preventing unacceptable radiation doses to workers and individuals of the public.

The hot-cell that is being implemented at CDTN, is shielded sufficiently for handling of ^{60}Co sources up to 7.4 GBq (200 mCi) and ^{137}Cs sources up to 740 GBq (20 Ci). The hot-cell include bricks and shot, radiation shielded windows, remote tongs, equipment mounting frames, a dose calibrator, etc. The design is presented in the figure below.

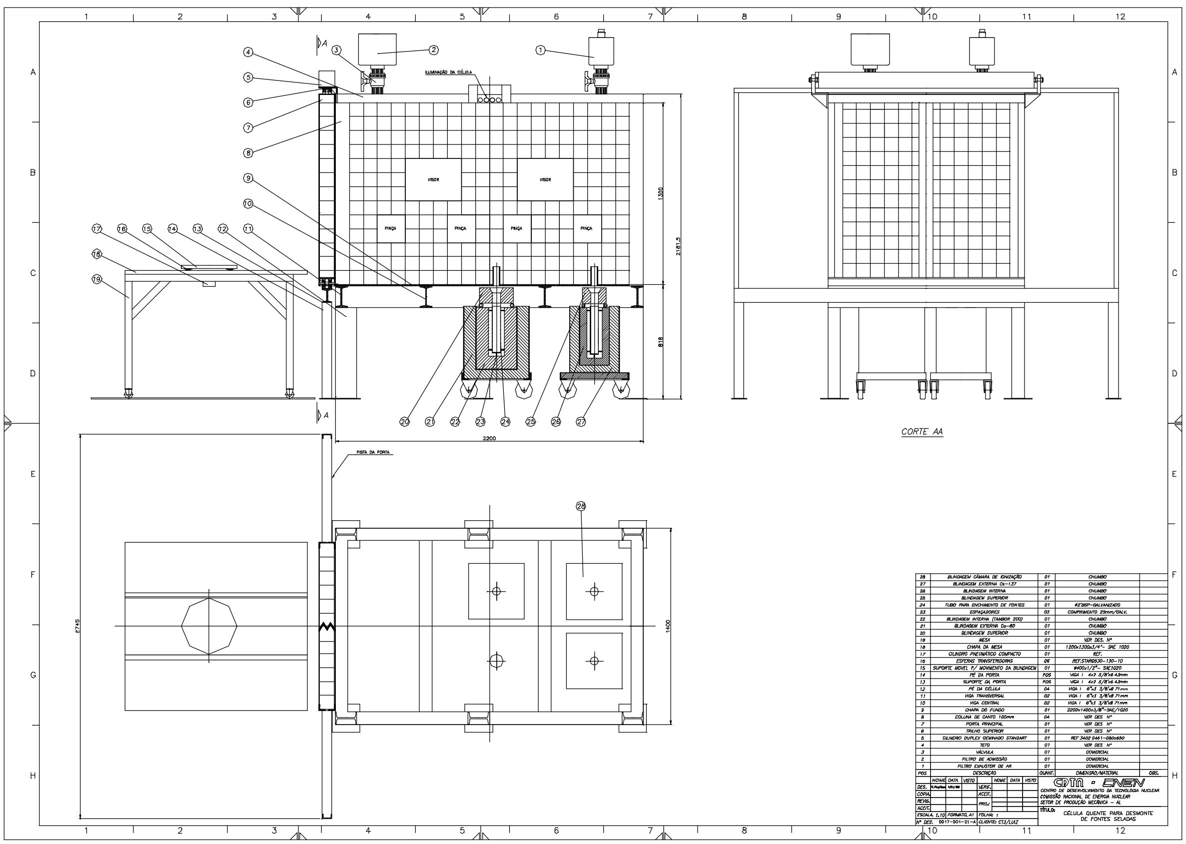

Fig. 3. Hot-cell for Dismantling Nuclear Gauges at CDTN

CONCLUSIONS

The application of the above methodology for conditioning of the existing ^{137}Cs sources at CDTN (78 units on 30/04/98) - will result in 8 packages, while the use of the methodology recommended by IAEA would generate about 39 drums (2 sources in each drum).

The conditioning of 109 sources of ^{60}Co will produce only one 200 L drum in contrast to the IAEA methodology, which would generate about 27 drums (4 sources in each drum).

The level of radiation on the surface of each drum was calculated with "Microshield". For 74 GBq of ^{60}Co the estimated radiation level was 0.890 mSv/h and for 370 GBq of ^{137}Cs the level was 0.887 mSv/h.

The same methodology will be applied for conditioning of other radioisotopes, reducing the number of conditioned drums from about 26 to 2 for these other radioisotopes.

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