## Essays on Nuclear Energy & Radioactive Waste Management

Ricardo Bastos Smith (Org.)



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### - 4 -Further Analyses of the Unburied Goiania Accident Packages<sup>6</sup>

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Abstract: In 1987, in the city of Goiania, Brazil, a derelict teletherapy machine was disassembled by scavengers and Cs-137 was released in the environment, unleashing the biggest radiological accident in Brazil. During the 15 days before the accident was acknowledged, some contaminated materials were sold and delivered to recycling factories in a few cities in the state of Sao Paulo, Brazil, in the form of metal scrap and recycled paper bales. The contaminated material was then collected, the metal scrap was conditioned in forty-three 200-liter drums, and the paper bales were stored in fifty 1.6 cubic meter steel boxes at the interim storage of the Nuclear and Energy Research Institute (IPEN), in the city of Sao Paulo, and there remained ever since. In 2017, 30 years later, initial analyses were performed at a sample of these boxes, checking for their activity, weight, and incongruences between the original values recorded at the time of collection and the measurement results 30 years later. The results indicated that none of the boxes checked were close to the clearance limit and that, without any sort of treatment, this radioactive waste should be stored for at least 150 years more. Visual inspection could not be performed at that time. Nowadays, some of the boxes were opened and samples from the contaminated material inside were taken for analysis. The main objective of this work is to report the results from the evaluation of the physical state of this material. After

<sup>6</sup> Lecture presented at the 2019 Waste Management Symposia (WMS) on March 03-07, 2019 in the city of Phoenix, AZ, United States. Available at: <a href="http://amz.xcdsystem.com/A464D2CF-E476-F46B-841E415B85C431CC\_finalpapers\_2019/FinalPaper\_19161\_0224053147.pdf">http://amz.xcdsystem.com/A464D2CF-E476-F46B-841E415B85C431CC\_finalpapers\_2019/FinalPaper\_19161\_0224053147.pdf</a>>.

these analyses, the treatment options for volume reduction that were previously proposed were reviewed, and the method that best suits the current characteristics of the waste was chosen.

Resumo: Em 1987, na cidade de Goiânia, Brasil, uma máguina de teleterapia abandonada foi desmontada por catadores e o Cs-137 foi lançado no meio ambiente, desencadeando o maior acidente radiológico do Brasil. Durante os 15 dias anteriores à descoberta do acidente, alguns materiais contaminados foram vendidos e entregues a fábricas de reciclagem em algumas cidades do estado de São Paulo, Brasil, na forma de sucata de metal e fardos de papel reciclado. O material contaminado foi então coletado, a sucata metálica acondicionada em guarenta e três tambores de 200 litros. e os fardos de papel armazenados em cinquenta caixas de aço de 1,6 metros cúbicos no depósito intermediário do Instituto de Pesquisas Energéticas e Nucleares (IPEN), na cidade de São Paulo, e lá permaneceram desde então. Em 2017, 30 anos depois, foram realizadas análises iniciais em uma amostra dessas caixas, verificando sua atividade, peso e incongruências entre os valores originais registrados no momento da coleta e os resultados da medição 30 anos depois. Os resultados indicaram que nenhuma das caixas marcadas estava próxima do limite de liberação e que, sem gualquer tipo de tratamento, esse rejeito radioativo deveria ser armazenado por pelo menos mais 150 anos. A inspeção visual não pôde ser realizada naquele momento. Atualmente, algumas das caixas foram abertas e amostras do material contaminado em seu interior foram retiradas para análise. O objetivo principal deste trabalho é relatar os resultados da avaliação do estado físico deste material. Após essas análises, as opções de tratamento para redução de volume propostas anteriormente foram revistas, e o método que melhor se adequa às características atuais do resíduo foi escolhido

#### Introduction

In 1987, a year and a half after the Chernobyl accident in the USSR, in the city of Goiania, Brazil, a teletherapy machine taken from a derelict radiotherapy clinic was disassembled by scavengers, and approximately 50.9 TBq of Cs-137 were released in the environment, unleashing the biggest radiological accident in Brazil [1].

The radioactive material, in the form of cesium chloride, was spread in the scrapyard, in a paper recycling company and among many individuals as well as their homes. Four persons died within a month after the accident, and a total of about 4.5 thousand tons of radioactive waste were collected during the cleanup operation, conditioned in boxes and disposed in a repository especially built for it, in the city of Abadia de Goiás, approximately 20km from the original contamination site [2].

Only 15 days after the beginning of the cesium dispersion was that the local sanitary vigilance acknowledged that some people were suffering from acute radiation syndrome and informed the authorities who proceeded on the identification and cleanup of the contaminated people and sites. During this time period, some contaminated materials were sold and delivered to recycling factories in the cities of São Paulo, Osasco, Araras and São Carlos, in the state of Sao Paulo, up to 1,000 km from the contamination site, in the form of metal scrap and recycled paper bales.

A new cleanup operation was performed. The contaminated material was collected; the paper bales were stored in fifty 1.6 cubic meter steel boxes, and the metal scrap was conditioned in forty-three 200-liter drums [3]. Because of the turmoil in the country and the sensibilization triggered by the radiological accident, as well as the transportation costs, it was not possible to deliver this waste to Abadia de Goiás. Therefore, it was brought to the interim storage of the Nuclear and Energy Research Institute (IPEN), in the city of Sao Paulo. The steel boxes and drums were there gathered in 1988, and besides the regular maintenances to fix scratches and corrosion points, the packages remained without any other verification until the year 2017.

With the 30 years of the greatest radiological accident in Brazil, the interest in analyzing this waste came up, aiming at comparing the old records with the current ones to be measured, and considering some methods for treatment of this waste in order to reduce its volume, as the project of a Brazilian radioactive waste disposal site is in progress, and in the coming years it will be necessary to transport this waste to its final destination.

Therefore, a sample of 14 boxes was selected, and measurements were performed in each box, from weight to dose rates at the sides and at different distances, to assess the heterogeneity of contaminated materials inside the boxes. The activities of each of the measured boxes were then calculated using the point-kernel method described by Rockwell, and the Microshield<sup>®7</sup> v.9.03 software package [4].

The results indicated that the estimated activities are in disagreement with those calculated 30 years before, and confirmed that part of this radioactive waste must be kept in storage for at least 150 years more, before reaching the clearance level. A few treatment methods were considered but only on a tentative basis, as it was not possible at the time to open any of the boxes and collect waste samples.

A new research was now performed aiming to conclude these analyses and determine the most appropriate treatment method for reducing the volume of this waste.

#### Methods

The objective of this research is to perform visual inspection of the waste inside the boxes, and to collect samples for laboratory analyses. Considering that no large enough cell with air containment is available and in order to avoid possible air contamination when opening the boxes, due to potential spreading of radioactive dust or microorganisms, a plastic cover that would serve as a containment (Figure 1) was assembled over the box.

<sup>7</sup> MicroShield® is a registered trademark of Grove Software, Inc.



Figure 1 - Plastic cap over the waste box.

The cover allows samples to be taken from the box without the risk of contamination of the air or surrounding surfaces. Gloves were installed on the front and lateral sides of the cover to allow unscrew the bolts and lift the lid. An acrylic plate was added to the front of the cover for better visual inspection. Such actions aimed at ensuring the environment was kept clean and safe.

Plastic bottles were left inside the containment before assembling for the collection of the samples.

Samples were analyzed with respect to activity concentration, pH, humidity, and the presence of microorganisms.

#### Results

After opening three boxes, the presence of high moisture content was observed (Figure 2), as well as a marked reduction in paper volume of the order of 10 to 30% (Figure 3).



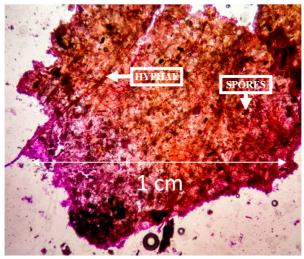
**Figure 2 -** Inside of the box that showed the highest moisture content. Note the water droplets formed by condensation in the underside of the box lid.



**Figure 3** - The paper bale collapsed, reducing the volume by about 30% of the original height. By the touch, the mass appears like moist clay.

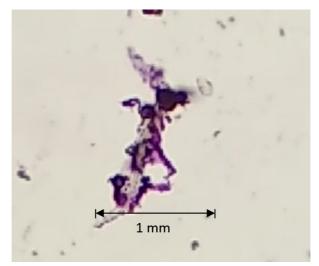
In 1988, the paper bales were filled up to the limit of the capacity of the boxes, as seen in page 45. Therefore, the volume reduction is associated with the moisture build up, which could only be explained by microbial action.

The paper bales, originally tied and wrapped in a plastic bag, have fallen apart and the paper degraded up to the point that the cellulose fibers appear broken in the examination under the microscope. The paper in all samples appeared as small fragments, visibly degraded [5], with colors varying from light brown to dark brown or black. In some parts of the box with the highest moisture content, it looked like a soft, wet mass, like moist clay.



**Figure 4 -** Stained sample of degraded paper showing the original cellulose fibers and the microbial mass with the hypha and spores of fungi.

Examination under the microscope (Nikon, Eclipse E600) showed that microbial life thrives in the waste mass. Figure 4 shows a sample stained with gentian violet (Hexamethylp-rosaniline chloride) and confirms the presence of fungi. Figure 5 shows a photogram, from a captured video of a living worm that was identified as a free-living nematode. It is certain that the bacteria are also present, but these microorganisms could not be identified in this examination of samples.



**Figure 5 -** A free-living nematode appears in this photogram of a video taken with a microscope from a stained sample of the paper mass.

Waste sample moisture content was measured with a moisture analyzer OHAUS, model MB200. Samples with approximately ten grams were kept at 100 oC until constant weight. TABLE I shows the results of both humidity and pH measurements.

The activity concentration of Cs-137 was also measured in samples of the three boxes, using passive gamma spectrometry, and the results are presented in TABLE II. Although the measurement of Box 350's sample may be already close to the discharge limits, further evaluations should be considered due to the high level of heterogeneity in the boxes, as explained by Tessaro, Geraldo, Souza, Smith and Vicente [4].

Sample box no.	Initial sample weight (g)	Final sample weight (g)	Moisture content (%)	Heating time (min)	рН
350	10.381	5.220	49.7	100	6
350	10.005	4.550	54.5	180	6
340	10.243	5.880	42.6	90	7
1334	9.777	5.020	48.7	70	7

Table I - Results of Moisture Content and pH Measurements

Table II - Activity Concentrations in the Samples of the Boxes

Sample box no.	Sample weight (g)	Radionuclide	Activity concentration (Bq.kg-1) (*)
350	37.12	Cs-137	19 ± 3
340	20.78	Cs-137	(2.24 ± 0.13) x 104
1334	21.19	Cs-137	(2.67 ± 0.13) x 106

(\*) Confidence interval:  $\pm 1 \sigma$  (68%)

### Conclusions

The unexpected presence of a high moisture content inside the waste boxes had two consequences: the first and immediate one was that the opening of the boxes did not require a containment to prevent air contamination by spreading of radioactive dust or microorganisms. Because of that, the containment hood was not used in the next two boxes that were opened.

The second consequence was that the original idea of using physical-chemical methods to treat the waste and reduce its volume was abandoned in exchange of a method using microorganisms which attack the complex lignin molecule, as paper may contain up to 20% of lignin [6]. The microbial action on the paper bales proved to be equally effective in reducing the volume of this waste and, in addition, has the advantage of being less aggressive to the metallic boxes.

This result is suggestive of conducting a pilot experiment in order to evaluate the feasibility of using especially selected microbiota to further reduce the volume of the waste. Time, cost, adverse factors and test running conditions, like temperature, humidity and the addition of chemicals that act as energy supply for the microorganisms are going to be used in the study design, prior to the performance of a full-scale treatment project.

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## Further Analyses of the Unburied Goiania Accident Packages

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