

WASTE VOLUME REDUCTION BY SPRAY DRYING

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ABSTRACT

The operation of nuclear facilities generates liquid wastes which require treatment to control the chemical compounds and removal of radioactive contaminants. These wastes can come from the cooling of the primary reactor system, from the reactor pool decontamination, washing of contaminated clothing, among others. The ion exchange resin constitutes the largest fraction of this waste, classified as low and intermediate level of radiation. According to CNEN Standard 8.01, the minimization of the volume and activity of the radioactive waste generated in the operation of a nuclear installation, radiative installation, industrial mining installation or radioactive waste deposit should be ensured. In addition, one of the acceptance criteria for wastes in repositories required by CNEN NN 6.09 is that it be solid or solidified. Thus, these wastes must be reduced in volume and solidified to meet the standards and the safety of the population and the environment. The objective of this work is to find a solution that associates the least generation of packaged waste and the acceptance criteria of waste for the deposition in the national repository. This work presents a proposal of reduction of the volume of the liquid wastes generated by nuclear facilities by drying by for reduction of volume for a greater incorporation of wastes in cement. Using spray dryer, an 18% reduction in the production of cemented waste products was observed in relation to the method currently used with compressive strength measurement above the standard, and it is believed that this value may increase in future tests.

1. INTRODUCTION

In Brazil, the generation of nuclear energy comes from the Angra 1 plants, with capacity of 640 megawatts, and Angra 2, with a capacity of 1,350 megawatts, operated by Eletrobrás Eletronuclear. Eletronuclear stores and controls full-time the wastes from Angra nuclear power plants, as well as a program for permanent monitoring of the levels of air, earth and water radiation around the Almirante Álvaro Alberto Nuclear Center (CNAAA). The radioactive wastes remain in deposits, within the CNAAA site, in Itaorna, until they will be transported to the repository, place destined to the final storage of the radioactive waste, after treated and conditioned [1].

At Angra power plants there are processes that generate liquid wastes at the nuclear facilities which require treatment to control chemical compounds and removal of radioactive contaminants. These wastes come from the cooling of the primary reactor system, from irradiated fuel pool decontamination, washing of contaminated clothing, among others. The

ion exchange resin is the main treatment for the removal of contaminants, and constitutes the largest fraction of the waste, classified as low and intermediate level of radiation.

According to the CNEN NN 8.01 standard, the minimization of the volume and activity of the radioactive waste generated in the operation of a nuclear installation, industrial mining installation or radioactive waste deposit should be ensured [2]. In addition, one of the acceptance criteria for wastes in repositories required by CNEN NN 6.09 is that it be solid or solidified [3]. Thus, these wastes must be reduced in volume and solidified to meet the standards and guarantee the safety of the population and the environment.

This paper describes the method used for volume reduction that is the drying process by atomization, carried out through a spray dryer. This process is disseminated by the process industry, in general, and it is nowadays specially applied for the large-scale drying of food, pharmaceutical products, among others [4].

The spray-drying process consisted in pumping the fluid product into the atomization chamber where it is atomized by subjecting it to a controlled stream of hot air, generating the evaporation of the solvents, obtaining the separation of solids and contained solids, with the minimum degradation of the product in drying, finishing the process with the recovery of the dry product [4].

This work presents a proposal of reduction of the volume of the liquid wastes generated by nuclear facilities by drying by for reduction of volume for a greater incorporation of wastes in cement. Using spray dryer, an 18% reduction in the production of cemented waste products was observed in relation to the method currently used with compressive strength measurement above the standard, and it is believed that this value may increase in future tests.

2. ACCEPTANCE CRITERIA IN BRAZIL FOR THE DISPOSAL OF LIQUID WASTES

The radioactive waste generated from the operation of the NPP is generally classified as low and intermediate level waste. In Brazil spent fuel from NPP is not considered radioactive waste and there is a pending technical, economic and political decision of the Federal Government about the possibility of reprocessing this fuel or disposing it of as such [5].

In Brazilian NPPs, Angra 1 and Angra 2 their waste is processed, stored, treated when necessary, and may be discharged in accordance to the safety rules established by nuclear and environmental authorities regulatory bodies (CNEN and IBAMA respectively). The solid waste should have its potential of migration and dispersion of radionuclides reduced, for this, the NPPs process the spent resin, the concentrated liquid was contaminated filters and the solid waste produced in the operation and maintenance of the plants, and confine them in special packages.

For the final disposal of low and intermediate-level waste, in Brazil is used the standard CNEN NN 6.09 [3], in which the waste criteria for safety disposal on repository are established, in order to ensure the protection for workers, population, and environment against the hazards of ionizing radiation.

Some of the general criteria established in this standard consist on knowing the content of radionuclides, the dose rate of the package, the traction and compressive strength, free liquid in the package, among others [3].

It is determined that all liquid wastes, resins, non-compressible solid wastes and filters should be embedded in solid matrix, homogeneously distributed and with the least segregated material, forming a monolithic.

3. WASTE TREATMENT METHODS

There are many different processes to treat liquid radioactive waste and it is important to know the factors to select the best process, as well know the possible processes.

3.1 – Important Factors for Process Selection

Considering the established criteria for the safe disposal of nuclear waste, is important to consider some factors that are relevant for selecting its treatment. These factors are the control of wastes at point of generation, their minimization and on, the level and nature of activity, their physical form, the secondary waste production, the physical chemical properties of the final waste, and the cost of treatment, storage and transport. Additionally it is necessary to take into consideration the transport requirements and the disposal system.

3.2 – Processes for Volume Reduction

There are a number of different processes which can be used for the treatment of radioactive waste, many of these dewatering and drying techniques has been applied in other fields such as food industry for years, thereby developing a secure, efficient and cost effectively technology. [6].

Since the legislation determines that the radioactive waste should be embedded in solid matrix, it is important to reduce the waste volume to improve the compatibility with immobilization processes.

3.2.1 – Forced Ventilation Oven

In convective drying using a forced ventilation oven, the material to be dried is placed in a container which is placed on a shelf within the oven. This stove is a thermal insulation chamber with heating system and circulating air ventilation. The main heating mechanism consists of the convection by the transfer of energy by the hot air stream in the closed environment [4]. Air is circulated on the surface of the product at a relatively high speed to increase the efficiency of heat transfer and the transfer of the material.

3.2.2 – Spray Drying

A method used for volume reduction is the atomization drying, carried out through a spray dryer. This process is disseminated by industries in general, and is mainly applied for the large-scale drying of food, pharmaceutical products, among others [4].

The spray-drying process consists in pumping the fluid product into the atomization chamber where it is atomized by subjecting it to a controlled stream of hot air, generating the evaporation of the solvents, and obtaining the dry product, as shown in the figure 1.

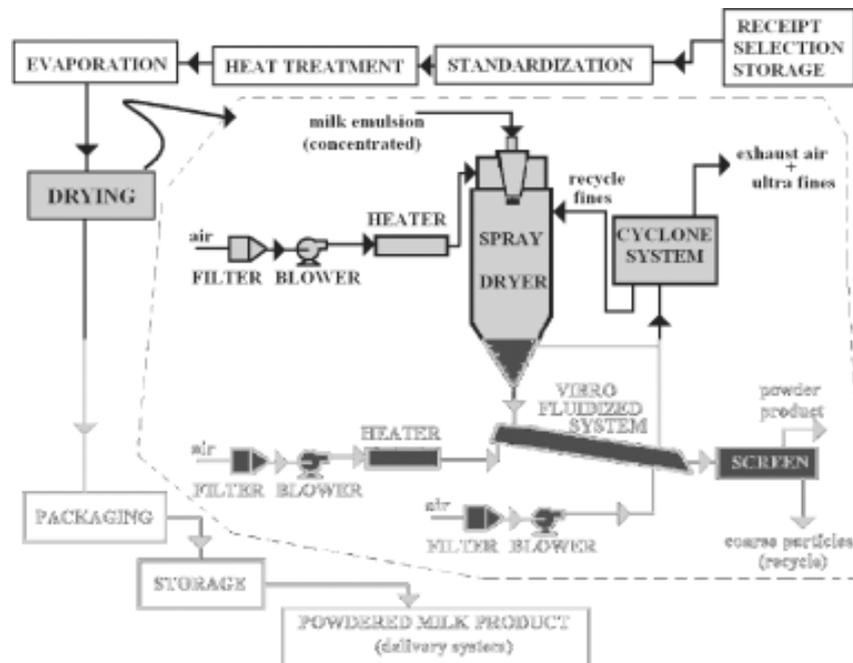


Figure 1: Spray Dryer Scheme [7]

Its effectiveness is based on the principle of increasing the area of contact between the material to be dried and the desiccant agent, in this case, hot air. For spheres, when their diameter is reduced by half, there is an eightfold increase in their surface area. This feature of atomizing a high surface area per gram of liquid is one of the main advantages of the spray dryer [8]. In addition to this advantage, the spray-drying process has a continuous operation and is adaptable to automatic controls, thus enabling process performance to be improved and done remotely [9].

However, this process has disadvantages, such as the high cost of installation and operation, non-viability for high density products and the product must be pumpable for the atomizer input [5].

3.3 – Cementation

In order to obtain a safer deposition of low and intermediate level waste, it is important that they be immobilized in order to reduce the potential migration or dispersion of radionuclides to the environment by natural processes during storage, transportation and final disposal, in addition to ensuring stability and durability.

Cement has been used as the main matrix to immobilize radioactive wastes, although bitumen and polymers are an alternative. Some of its advantages are, the material and technology are well known, it is compatible with many types of wastes, most liquid wastes bind chemically to

it, the low cost of cement, good shielding ability, good compressive and impact strength, low leachability for various nuclides, no free water when well formulated, and fast controlled hardening, without precipitation or segregation during curing [10]. However, other matrices are used according to the situation, such as the type and shape of the waste, activity and radionuclides present, requirements of the repository, as well as economic, engineering and safety factors.

4. METHODOLOGY

4.1 – Materials and Equipment

4.1.1 – Materials

For the preparation of the blends and the molded cement cylinders were used the following materials: Portland cement (CP V-ARI); Deionized Water; Calcium Hydroxide; Sodium Hydroxide; Superplasticizer Sika® Viscocrete® 20 HE; Active silica; Brasgel Bentonite Clay; Simulated Waste.

For the preparation of the simulated waste, was used the ion exchange resin Amberlit IRN217® (immersed in water);

4.1.2 – Equipments/Instruments

The equipments and instruments used on the preparation of pastes, molding of the cement cylinders and the physico-chemical tests were the following: Caliper, model Universal, n° de série 530.104, Mitutoyo; Analytical balance Shimadzu; Balance Balmak ELP-10; Stand mixer Arno Ciranda 110V; Steel molds with 5cm diameter (adjustable) per 10cm; Ph Indicator strip Merck Neutralit pH 5-10; Digital chronometer Technos, model 219; Hydraulic press EMIC PCI 150/200, indication range 30.000 kgf and scale division 1 kgf.; Analytical digital balance Gehaka, model BG8000; Pilot Spray Dryer APV-ANHIDRO PSD model 52; Precision lab oven 200°C Sterilife – forced air circulation.

4.1.3 – Spray dryer

The drying of the resin by spray dryer was conducted in a pilot spray dryer, Figure 3, with drying chamber of 1m in diameter and evaporation capacity of 7.5 kg/h. The equipment uses a centrifugal-type sample atomizer system with rotary disc, positioned in the center of the air diffusers, giving an air-spray contact. The dry material was collected in two parts: the heavy ones by gravity in a bottom collector and the fines ones separated in cyclone, all collected material was homogenized.



Figure 3: Spray Dryer APV – IEN-RJ.

The resin was inserted into the equipment by metering pump at the flow rate of 40 mL/min while being mixed in a shaker to homogenize the sample at room temperature, the inlet air temperature was 150°C.

4.2 – Cementation test with ion exchange resin

4.2.1 – Preparation of pastes and molding

In the following to the humidity reduction methods, cementation tests were performed. The formulation and the procedure used for the cementation of the wet resin followed these steps:

- a) Weigh the separated material in the amount specified in Table 2;

Table 2 – Material quantities for the resin test

Material	Quantity (g)
Clay	9,1
Cement	416,0
Ca(OH) ₂	9,1
Active Silica	41,6
Superplasticizer	3 + 2,3
Resin	225

- b) Mix the active silica with cement, homogenize and separate into three parts (Table 3);

Table 3 – Quantity of cement and active silica mix for the resin test

1 st Part (g)	2 nd Part (g)	3 rd Part (g)
153,6	152,0	152,0

- c) Transfer the loaded resin into the container where the mixture will be held, turn on the mixer and add water;
- d) Add Ca(OH)₂ and measure the pH of the neutralized resin;
- e) Add the clay, the 1st part of the cement + active silica mixture (Table 3), and the 1st part of the superplasticizer;
- f) Add the 2nd part of the cement + active silica mixture (Table 3), and the 2nd part of the superplasticizer;
- g) And, add the 3rd part of the cement + active silica mixture (Table 3) and mix the paste;
- h) Mold the test specimens on cylinder moulds, seal the cylinders and store them for 28 days.

For the cementation of the dry resin by spray dryer, the steps used were the same, with change in the amount of water and resin added in the process, according to Table 4.

Table 4 – Amount of resin and water added in the dry resin cementation process

Resin (g)	Water (g)	Total (g)
67,00	193,52	260,52
82,00	193,52	275,52

4.2.2 – Compressive strength measurement

An important property of the monoliths is the compressive strength, which ensures the integrity of the product in the case of accidents involving a mechanical impact. This test consists of subjecting test bodies to increasing and continuous tension until their deformation or breakage, performed as shown in Figure 2. It is directly related to the homogeneity of the monolith, amount of pores and the presence of cracks in the solid [11].



Figure 2: Compressive Strength Test

The main purpose of the test is to evaluate the resistance of the product from impacts, especially during handling and transportation. The compression strength test for Portland cement is defined and based on standard NBR 7215 [12].

After 28 days of curing was evaluated the mechanical strength of the test specimens (Figure 4) produced by compressive strength test.



Figure 4: 28 days curing specimen.

To perform the test, the molded specimens were adequate, sanding their bases so that the force applied by the press was homogeneous across the surface and measured their heights, radii and

masses. After preparation, the samples were placed in the press for the measure of its compressive strength

5. RESULTS AND DISCUSSION

After the compressive strength tests were carried out, the results presented in Table 5 were obtained, in which the specimens R1 and R2 are the dry samples by laboratory oven, with 67 and 82g, respectively, and the specimens SD1 and SD2 are the dry samples by spray dryer, with 67 and 82g, respectively of incorporated resin.

Table 5 – Results of the compressive strength tests of dry resin cementation

Sample	Diameter (cm)	Height (cm)	Weight (g)	Force (kgf)	Pressure (MPa)
R1A	49,9	100,68	329,9	1697	8,57
R1B	49,81	100,55	329,6	2980	14,99
R2A	99,6	99,84	321,1	2735	13,75
R2B	49,9	101,05	325,7	3263	16,36
SD1A	50,06	100,30	323,8	2382	11,99
SD1B	50,23	101,44	325,0	2342	11,78
SD2A	49,83	100,67	332,8	3120	15,52
SD2B	49,80	99,71	329,6	2575	12,76

Evaluating the results, it can be observed that the result of the compressive strength of the specimens with dry resin in oven and spray dryer showed a value above that required by the standard, 10MPa, with the exception of only one body, R1A. The best strength values are of the specimens with a higher amount of dry resin incorporated.

6. CONCLUSIONS

From the tests and studies carried out in this work, it is possible to affirm that the drying of the waste is an efficient method to reduce the volume of cemented wastes.

The amount of water/resin in the solution used for current cementation is 225g, these tests showed that a raise of the solution to 260,52g and 275,52g is possible by the drying procedure, and that this amount can still be optimized to increase the incorporation.

By drying the ion exchange resin, there is a total reduction of 18% cemented products. This reduction of cemented products promotes better management in the space that the waste will occupy in the repository provided by the RBMN project, thus making the repository more efficient. In addition to reducing transportation and cargo risks for future generations.

And because spray dryer is a widely used technique in many industries, its application in the nuclear area would be an easy transition process, proving to be a viable option with benefits and advantages that outweigh the disadvantages and other techniques.

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REFERENCES

1. “ELETROBRAS ELETRONUCLEAR” <http://www.eletronuclear.gov.br> (2014);
2. CNEN – COMISSÃO NACIONAL DE ENERGIA NUCLEAR. “Gerência de Rejeitos Radioativos em Instalações Radiativas”, *CNEN-NE-8.01*, Rio de Janeiro, (2014);
3. CNEN – COMISSÃO NACIONAL DE ENERGIA NUCLEAR. “Critérios de aceitação para deposição de rejeitos radioativos de baixo e médio níveis de radiação”, *CNEN-NN-6.09*, Rio de Janeiro: CNEN, (2002);
4. A. S. MUJUMDAR, *Handbook of Industrial Drying*, 3rd edition, Singapura, (2006);
5. REPÚBLICA FEDERATIVA DO BRASIL. “National Report of Brazil 2014 for the 5th Review Meeting of the Joint Convention on the Safety of Spent Fuel Management and on the Safety of Radioactive Waste Management”, Rio de Janeiro, Brazil, (2014);
6. B. ZEKI. *Food Process Engineering and Technology*, Academic Press, Burlington, United States of America, (2009);
7. V. S. BIRCHAL; M. L. PASSOS. Modeling and simulation of milk emulsion drying in spray dryers. *Brazilian Journal of Chemical Engineering*, **vol.22** n°2, São Paulo, (2005);
8. E. D. ROSA; M. TSUKADA; L. A. P. FREITAS. Secagem por atomização na indústria alimentícia: fundamentos e aplicações. *Jornada Científica Da Fazu/Faculdades Associadas De Uberaba*, vol.5, Uberaba, (2006);
9. R. P. PATEL; M. P. PATEL; A. M. SUTHAR. Spray drying technology: an overview. *Indian Journal of Science and Technology*, **vol.2**, p.45-52, (2009);
10. IAEA - INTERNATIONAL ATOMIC ENERGY AGENCY. *Report on radioactive waste disposal (Technical Reports Series N° 349)*. Vienna, (1993);
11. P. K. MEHTA; P. J. M. MONTEIRO. *Concreto - Estrutura, Propriedades e Materiais*. Pini, São Paulo, (1994);

12. ASSOCIAÇÃO BRASILEIRA DE NORMAS TÉCNICAS. *ABNT-NBR 7215. Cimento Portland: Determinação da Resistência à compressão*. ABNT, Rio de Janeiro, (1991);