BJRS

BRAZILIAN JOURNAL
OF
RADIATION SCIENCES
09-01A (2021) 01-14

Immobilization of liquid radioactive waste in cement

França[1] A. A. S., Vicente[1] R.

[1] Nuclear and Energy Research Institute (IPEN/CNEN - SP)

Professor Lineu Prestes Avenue 2242

05508-000 São Paulo, SP, Brazil

profandreza.franca@gmail.com

ABSTRACT

Immobilization of radioactive waste is required to comply with nuclear regulations and waste acceptance criteria in a repository, which require the waste to be solid or immobilized in solid form within a durable and resistant matrix [1-4]. Cement is the most frequently used material for the immobilization of liquid, low-level waste, since it has many advantages, such as the ease of preparation at room temperature and the low cost [5, 6]. In this paper, we describe the characteristics of cement-water mixtures, homogenized in a drum using a vibration table as the mixing device. Common Portland cement was used as the immobilization matrix. The homogeneity of the mixtures is evaluated using cement dye in appropriate amounts. Initially, the distribution of the mineral dye was made by visual inspection. The batches were carried out with three different ways of feeding the components. Different results were obtained depending on the feeding methods employed.

Keywords: radioactive waste, mixing, cement.

ISSN: 2319-0612
Accepted: 2020-07-06

1. INTRODUCTION

According to VICENTE (2016), cementation is one of the most used methods to immobilize several types of radioactive waste. Immobilization is required to comply with nuclear regulations and repository acceptance criteria, which require the waste to be in solid form within a durable and resistant matrix [7].

The main advantages of cement as an immobilization matrix are:

• The process of mixing the waste with the matrix is simple and operates at room temperature;
• The technology is well demonstrated;
• The product is incombustible and thermally stable over a wide range of temperatures;
• The product has great chemical and biochemical stability;
• The waste can be incorporated or encapsulated in the matrix;
• The water contained in various types of waste is used in the cementation process.

The cementation process, that is, the mixture of waste, water and anhydrous cement, in order to hydrate and immobilize the waste, forming a solid monolithic block, can be carried out in several types of equipment, classified into three groups (VICENTE, 2016):

a) mixing in the final container itself, known as 'in-drum mixing'; b) batch mixing; c) continuous mixing.

1.1 In Drum Mixing

The first group is characterized by a system with one or two vertical axis mixers, with helical fins and planetary movement, which is lowered into the package and which promotes mixing and homogenization of the product. Figure 1 is a basic flow diagram of the process and an example of this type of equipment, the blender of the American company, Westinghouse Electric Co., with two axles, and a drum with the product just after mixing [8].

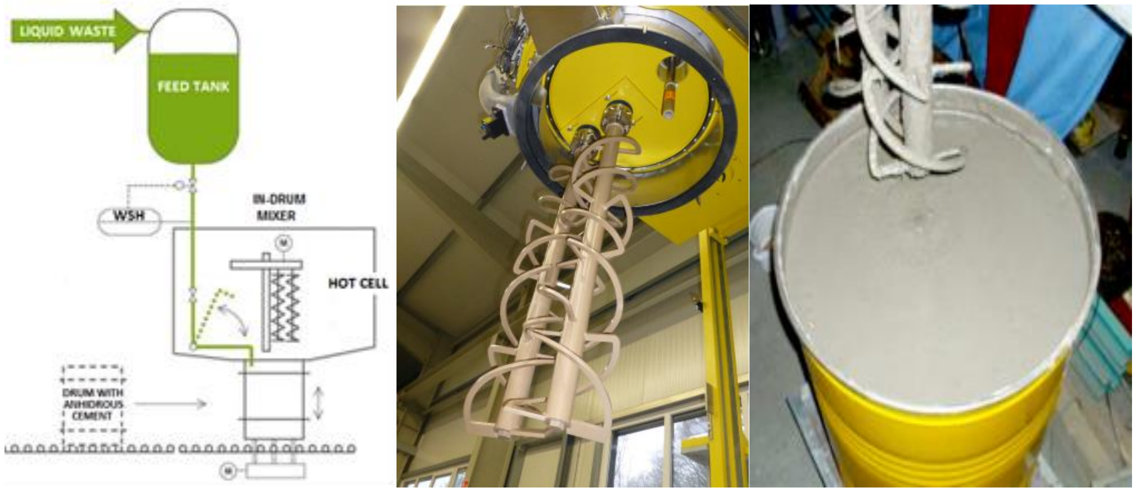


Figure 1: *In drum mixing process and Westinghouse Electric Co. cement mixer [8]***.**
Source: Westinghouse, 2015.

This group may be further divided into equipment with a) reusable mixers, and b) lost mixers. The mixer shown in figure 1 is reusable, while the lost mixers remain immersed in the mixture after the homogenization has been completed. The advantage of the former is that there is no need to replace the mixer with each package produced. The advantage of the second is that no cleaning of the mixer is necessary, to remove the residue of cement paste, which generates a secondary radioactive waste. The drawbacks of each are apparent from the advantages of the competitor, but there is still the problem of the volume of the blender that represents a loss of capacity of the package by leaving an empty volume at the top, or the presence of a 'foreign body' in the final product in the case of the lost blender.

The immobilization process is performed with a drum previously loaded with the amount of anhydrous cement necessary to immobilize the waste and the appropriate additives for the recipe. A conveyor and an elevator position the drum and attach it to an airtight box, which is a glovebox or a hot cell with tele manipulators, depending on whether the activity of the waste requires shielding or not. The waste is slowly poured into the drum while the already-started mixer is lowered until it reaches the bottom of the drum and promotes mixing. After the amount of waste previously determined is added and homogenized, the mixer is raised and vibrated so that most of the adhered cement paste flows into the drum and is then washed in a separate container. The lost mixer is

ISSN: 2319-0612
Accepted: 2020-07-06

disconnected from the driver and left inside the mixture. The quality control measurements are taken, the drum is closed, and dispatched to the radioactive waste storage.

1.2. Mixing in Batches

The second group is characterized by a fixed equipment, within which the waste is mixed with the cement and additives and then poured into the packages to solidify. All components of the blend are pre-dosed, and can be fed simultaneously or not in the mixer. The capacity of the mixer is high enough for one batch to fill several drums. The advantages of this equipment are that the cleaning of the parts that remain in contact with the radioactive paste need only be done once after several packages have been produced and final containers of any size and geometry can be used. In general, cement setting retarders are used so that the blend can flow easily until the last package is full. Figure 2 is an example of a flow chart that uses this type of equipment

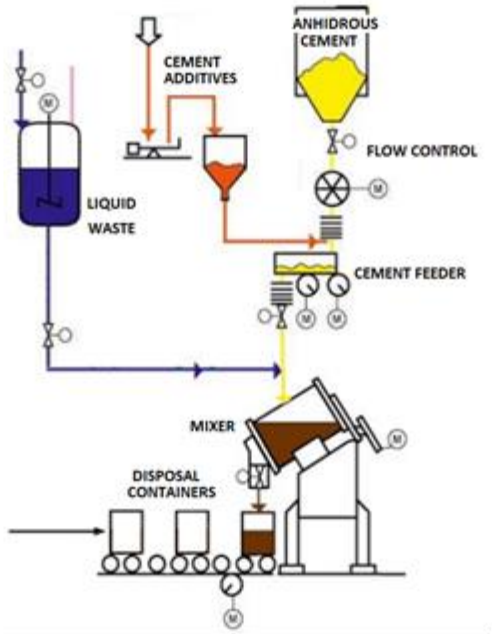


Figure 2: *Batch cementing equipment.*

ISSN: 2319-0612
Accepted: 2020-07-06

1.3. Continuous Mixers

A conveyor-type mixer that continuously mixes the components and, at the same time, transports the mixture to the point where it is poured into the final packages characterizes the third group of equipment. The great advantages of such equipment are that large quantities of waste can be immobilized with a minimum of process preparation work and there are no limits on the size of the final packages. Figure 3 is the flow diagram of the process.

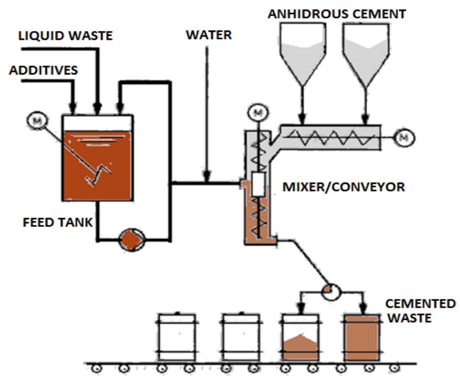


Figure 3: Flowchart of the continuous cementation process.

All these cementing processes present one or more disadvantages of cleaning the equipment, which generates a secondary liquid radioactive waste to be treated and, in the case of the 'in-drum' mixing, has the handling of the powdered cement in a in which the control of radioactive contamination requires cleaning. In the variants of the continuous process or batch mixing there is still a risk that the paste will harden and damage the equipment in case of any stop making the process stop (VICENTE, 2016).

A process of mixing the cement with the tailings to dispense moving parts of the equipment into contact with the cement paste has been tested on a small scale in the Laboratory of Radioactive Waste Management (GRR) of the Nuclear and Energy Research Institute, success, generating a homogeneous product, without causing the inconveniences already pointed out.

In the process tested in the GRR, the liquid waste is injected into the final package simultaneously to feed the cement, both of them under tightness and with the equipment connected

ISSN: 2319-0612
Accepted: 2020-07-06

to a compact exhaust and air filtration system. Except for the packaging, the exhaust duct and the filter, the pieces of equipment in contact with the tailings are not exposed to the cement powder and vice versa.

They compete to promote mixing and homogenization, simultaneous feeding and vibration of the package by means of a vibrator with circular movement in a plane inclined to the axis of the package.

According to VICENTE (2016), there are no mixers that are lost or need to be washed, to remove the cement paste, or to be decontaminated. The maximum capacity of the final package is achieved without leaving any voids at the top. If the blending and homogenization process is interrupted at some point it can be resumed at any time, without prejudice to damage of parts of the equipment, since none is in contact with the hardened cement paste.

2. MATERIALS AND METHODS

Vibrators with different actuating devices (electromagnetic and electromechanical) and with different movements (linear vertical or horizontal and circular bi- or three-dimensional) will be tested with different proportions and feed rates of the components of the mixture and different times of homogenization.

Common Portland cement, LIZ brand, CP IV-32 RS (ABNT NBR 5736 and NBR 5737) was used as immobilization matrix. Commercial product, newly produced and acquired from distributors of cement, was acquired from the local market.

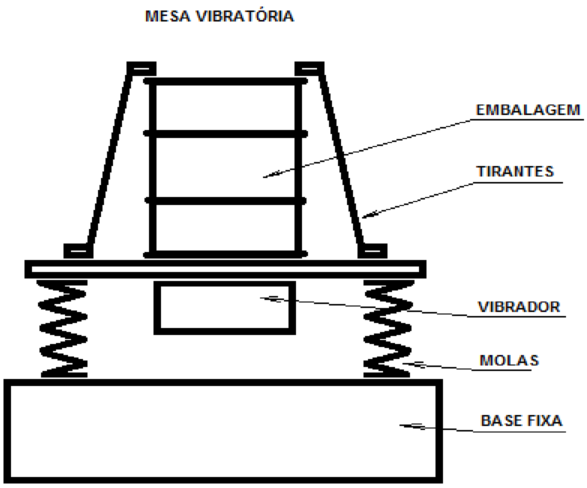
The homogeneity of the blend was evaluated with LANXESS iron oxide-based pigment powder in the blue color in quantities suitable for trace concentration measurement methods. In this case, the measurement of the mineral dye will be done by visual inspection or another method that is available in IPEN for samples of hydrated cement.

Samples for analysis of tracer concentration were withdrawn immediately after completion of the homogenization process in sufficient quantity to measure the homogeneity of the dye in the blend.

ISSN: 2319-0612
Accepted: 2020-07-06

The homogenization tests were carried out with the 3.5 L packages fixed by means of cable tensioners, shown in detail in figure 4, with screws of 3 mm to the vibration table, constituted by a platform that on its lower surface is fixed the vibrator device of the brand NORMA automaker Ltda., model VM-033, series 8533-6, 1750 RPM, 220/380/440 Volts, 0,33 HP and with unbalance of 30º, connected to an inverter of frequency of the WEG brand, model CFW10 at 15 Hz, supported by 16 compression springs of carbon steel with external diameter of 1.9 cm and length of 4.7 cm, anchored to a fixed base. Figure 5 is a schematic drawing of the assembly.

Figure 4: *In particular, cable tensioner used to secure the barrel*



ISSN: 2319-0612
Accepted: 2020-07-06

Figure 5: *Schematic drawing of a vibratory table for the cementation tests.*

For analysis, it was necessary to determine the amount of the reagents. Knowing that the capacity of the drum is 3.5 L, the water/cement ratio, by mass, used was 0.35 and the specific mass of the paste (cement and water mixture) is equal to 2 g/cm³.

V = 3,5 L

ρ_{paste} = 2 g/cm³ = 2 kg/dm³ = 2 kg/L

M_{paste}= 2 kg/L x 3,5 L = 7 kg

$$Mpaste = Mwater + Mciment = 7 \quad (1)$$

$$\frac{Mwater}{Mciment} = 0{,}35 \quad (2)$$

$$Mwater = 0{,}35\ x\ Mciment \quad (3)$$

$$0{,}35\ x\ Mciment + Mciment = 7 \quad (4)$$

$$1{,}35\ x\ Mciment = 7 \quad (5)$$

$$Mciment = \frac{7}{1{,}35} \quad (6)$$

$$Mwater = \frac{7}{1{,}35}\ x\ 0{,}35 \quad (7)$$

On what:

V = volume

ρ = density

M = Mass.

Each batch lasted a total of 25 minutes, with 20 minutes of food available, summarizing 1 feed per minute, totaling 20 feeding stages.

The following calculations were used to determine the number of masses per feed:

$$V\ ciment = \frac{7\ Kg}{1{,}35}\ x\ \frac{1}{20\ min} = 0{,}259\ \text{Kg/min} = 259\text{g/min} \quad (8)$$

ISSN: 2319-0612
Accepted: 2020-07-06

$$V\ water = \frac{7\ Kg}{1{,}35} x 0{,}35 x \frac{1}{20\ min} x \frac{1L}{1Kg} = \frac{0{,}090L}{min} = 90\ mL/min$$

(9)

Where V = Volume.

A precision scale of MARTE, model 1020 was used to prepare the samples. These samples were collected in disposable polypropylene (PP) cups of 180 ml.

2.1 Experimental Procedures

The tests were carried out with 3 different feeding methods: the first method consisted in feeding the batch with 359 g of cement and 90 ml of water every minute, being placed at the same time, totaling 20 stages and allowing to mix in the remaining 5 minutes, totaling 25 minutes mixing; the second method was based on feeding the cement rapidly while the feed of the water was made slowly until the next minute and hence the next feed of cement and allowing to mix in the remaining 5 minutes, totaling 20 feed stages; the third method consisted of starting with the drum containing all the water and making the 20 feeds with the cement every minute leaving the 5 minutes of mixing, totaling 25 minutes.

At the end of the tests, four test specimens were collected using a one-sided sampling probe for observation.

The increase of the scale will be done experimentally, starting the process of mixing with 3.5 L packs, then with 20 L packs and finally with 200 L drums.

3. RESULTS AND DISCUSSION

Different results were obtained according to the feed methods used.

In the first method, there was no homogeneity of the mixture, as no dye uniformity was observed and, at some points, cement powder was still not mixed with the water, shown in figure 6. It was noted that the water did not was absorbed by the cement, which accumulated in the center of the drum. In this way this method will no longer be used

ISSN: 2319-0612
Accepted: 2020-07-06

Figure 6: *First sample at end of test.*

In Figure 7, the specimens of the first sample.

Figure 7: *Bodies of proof of the first sampling.*

ISSN: 2319-0612
Accepted: 2020-07-06

In the second method, it was not observed homogeneity of the mixture and it is still not noticed uniformity of the dye, however the cement dots were still powdered without mixing with the water, as can be seen in figure 8. And we believe there was more homogeneity due to the way of feeding, which would be a point of difficulty in a larger scale process.

Figure 8: *Second sample at end of test.*

In Figure 9, the specimens of the second sample.

Figure 9: *Proof of second sampling.*

ISSN: 2319-0612
Accepted: 2020-07-06

In the third method, it was observed that there was a better mixing compared to the previous tests, however, cement spots still do not mix with the water, as we can see in figure 10. However, this method is more like industrial scale use, since the reject will already be in the drum and we will only add the cement. It was noted that to some extent the water was mixed with the cement until the viscosity of the fluid increased to the point that there was no more mixing with the frequency we were testing.

Figure 10*: Third sample at end of test.*

In Figure 11, the specimens of the third sample:

ISSN: 2319-0612
Accepted: 2020-07-06

Figure 11: *Proof of third sampling.*

4. CONCLUSIONS

The results are still inconclusive due to the lack of instruments needed to measure parameters such as frequency and range of motion, requiring new approaches and data analysis parameters.

In order to obtain better results, we will follow the tests by altering the test conditions, being it can be as much in relation to the power of the vibrating device as in relation to the frequency and amplitude of movement, as well as modifying the forms of feeding of the process, and, for changing the attachment of the vibrator to the table. The axis of the vibrator will be inclined relative to the plane of the vibratory table so that other vibration planes are induced in the mixing vessel.

We will proceed to the tests with the third method, because we obtained the best results, and many points were observed to improve, for example, the change of the vibratory frequency of the equipment depending on the viscosity of the mixture.

ISSN: 2319-0612
Accepted: 2020-07-06

5. REFERENCES

[1] INTERNATIONAL ATOMIC ENERGY AGENCY- IAEA, Vienna. "Conditioning of low- and intermediate- level radioactive wastes". Vienna, 1983. (IAEA Technical Report Series nº 222).

[2] INTERNATIONAL ATOMIC ENERGY AGENCY- IAEA," Classification of radioactive waste – General safety guide"**.** Vienna, 2009.

[3] COMISSÃO NACIONAL DE ENERGIA NUCLEAR- CNEN, "Gerência de Rejeitos Radioativos em Instalações Radiativas" – CNEN-NE-8.05, Rio de Janeiro, 1985.

[4] COMISSÃO NACIONAL DE ENERGIA NUCLEAR- CNEN, "Gerência de Rejeitos de Baixo e Médio Níveis de Radiação" – CNEN-NN-8.01, Rio de Janeiro, 2014.

[5] AMERICAN SOCIETY FOR TESTING AND MATERIALS- ASTM. *Standard guide for sampling strategies for heterogeneous wastes*. Designation: D5956−15, 2015.

[6] C. L. Grants and P. A. "Pelton Role of Homogeneity in Powder Sampling". ASTM, 1973. Disponível em: <www.astm.org>, Access at 21/08/17.

[7] VICENTE, R. "Projeto de Pesquisa: Processo para imobilização de rejeitos radioativos em cimento". IPEN, São Paulo, 2016.

[8] WESTINGHOUSE ELETRIC CO. "Decontamination, decommissioning and remediation / Global Waste Management", 2015. Disponível em: <www.westinghousenuclear.com>, Access at 25/07/2017.

ISSN: 2319-0612
Accepted: 2020-07-06