Evaluation of the Properties of Cement Pastes and Mortars Used in the Encapsulation of Bituminized Waste to be Disposed in a Near-Surface Repository

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Abstract: The Brazilian repository is being planned to be a near-surface one. In Brazil, the low and intermediate level radioactive wastes are immobilized using cement and bitumen for nuclear plant Angra 1 and Angra 2, respectively. The major problems due to the disposal of bituminized wastes in repositories are swelling of the waste products and their degradation in the long term. To accommodate the swelling of the bituminized wastes, the drums are filled up to 70-90% of their volume, which reduces the structural the repository stability and the disposal availability. Countries, which use bitumen in the solidification of NPP's radioactive waste and have near-surface repositories, need to immobilize this bituminized waste within other drums containing cement pastes or mortars to disposal them. This study aims to evaluate the properties of bitumen, cement pastes and mortars to be used in the encapsulation of bituminized wastes. The formulations of two pastes and two mortars were selected for the pilot scale tests. The laboratory and pilot scales results were very similar, indicating that any of these formulations could be used. However, the better formulation will be chosen after the leaching test results, because it is an essential parameter in the long-term repository performance.

Key words: Radioactive wastes, bitumen, cement pastes, mortars.

1. Introduction

In Brazil, there are currently two nuclear power plants with PWR (Pressurized Water Reactor) type in operation, located in Itaorna, Rio de Janeiro: Angra 1, Angra 2 and Angra 3 under construction. Angra 1 has the capacity to generate 640 MW and Angra 2, 1,350 MW. Angra 3 is expected to generate 1,405 MW [1].

In the Brazilian Nuclear Program was predicted the construction of, at least, four nuclear power plants in the Northeast and Southeast regions of Brazil, until 2030, to meet the increased use of nuclear power to generate electricity [1, 2]. With this growth and the increase of the use of radionuclides in different areas, the issue of radioactive waste deserves government actions that are been taken to resolve issues related to their storage [3].

Brazilian Federal Law 10.308 regulates the final destination of the radioactive waste produced in the country, including site selection, construction, licensing, operation, supervision, costs, damages, liability and warranties relating to deposits of radioactive waste. The responsibility of CNEN (National Commission for Nuclear Energy) for the final destination of the waste is established in the Article 2. In accordance with this regulation, it is not allowed receiving radioactive waste in liquid or gaseous forms in final repositories [4]. Therefore, the solidification of waste is required in order to meet three basic criteria to ensure safe handling of radioactive waste in repositories, which are mechanical resistance, permeability and stability. The matrices used are cement, polymers, glass and bitumen [5].

The immobilization and solidification of radioactive waste of low and intermediate level radiation (concentrated evaporated and ion exchange resins

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exhausted) from Angra 1 and Angra 2 are carried out in cement and bitumen respectively. It is planned to use bitumen for the immobilization of wastes Angra 3 [6].

The encapsulation of radioactive waste in bitumen is also used in Romania, Slovakia and the Czech Republic and in a nuclear power plant in Switzerland. This process was first used in some countries such as France, Belgium and Spain, and it was planned to be the bitumization used in a new nuclear power plant in Argentina. However, these countries abandoned and replaced with other processes, after the evaluation of the total cycle of waste from its generation to its disposal [7].

In general, most of the waste generated is low and intermediate level of radiation. The most common method of final disposition (disposal) of this type of waste is its confinement in near surface repositories (Fig. 1). The proposal of the Nuclear Energy Agency is to establish benchmarks and provide generic guidance on the methodology that can be used by national authorities to establish acceptance criteria for the specific location of the repository. Define three types of criteria:

- Limits on the concentration of radionuclides in the waste;

- Limit the total activity of radionuclides to be disposed in a given installation;

- Performance standard for packaged waste and packaging [8].

Due to the characteristics of near surface repositories the bituminized waste cannot be stored directly, but requires additional actions to ensure safe storage of these wastes and the security of the entire facility [9].

Experiences of bituminized waste disposal on surface repository were reported in countries like Romania, where ion exchange resins are encapsulated in bitumen in 60 liter drums, which are then packed in drums 200 L with an intermediate concrete shielding. Slovakia radioactive waste are incorporated into bitumen and this mixture is stored in drums 200 L coated with zinc (galvanizing), deposited in containers and transported to the surface Mochovce repository. In the Czech Republic, 100 L waste drums are encapsulated by high strength concrete in 200 L drums, plates and galvanized corrosion-resistant paint, and then sent to the repository Dukovany [7]. Other countries such as France and Belgium have opted for bituminized waste disposal on deep geological repository, which minimizes the risks associated with these wastes [9].

Fig. 1 Near surface repository.

Studies indicate serious concerns about the risks associated with bituminization radioactive waste, related to the process as well to the product. There are also major problems due to the presence of bituminization products in repositories, which are swelling of the product and waste. Long term degradation is caused by fissures, softened by temperature variations, creep and because of its visco plastic material, attacked by micro-organisms and, under certain conditions, fire hazard. To accommodate swelling, the drum must be filled in the range of 70-90% of its volume, which can decrease the structural stability of the repository and increase the area necessary for disposal [7].

The “RBMN Project” was launched in November 2008 and aimed to implement the Brazilian Repository, from its conception to its construction. The concept adopted is a surface repository constructed according to the inventory of radioactive wastes, including those from the operation of nuclear facilities, from the facilities of the nuclear fuel cycle, and from the use of radionuclides in medicine, industry and research activities in Brazil. The implementation of the national repository is an important technical requirement and currently a legal requirement for the entry into operation of the plant Angra 3 [3].

Most of the waste to be disposed in the repository will be from nuclear power plants. The acceptance requirements for disposal of these wastes are directly linked to the security of the entire installation. Therefore, a solution for the storage of bituminized waste must be obtained to ensure the performance expected by the safety analysis.

This study aims to look for solutions to dispose the bituminized waste products in the repository, making them compatible with the acceptance criteria of cemented waste products.

2. Material and Methods

According to international experience, two

packaging options for bituminized waste product will be selected, using bitumen encapsulated in cement (paste or mortar). The properties of the bitumen (without waste) used in Angra 2 and cement pastes (cement and water) and mortars (cement, water and sand) were tested.

2.1 Evaluation of the Properties of Bitumen

The tests conducted for this evaluation of the properties of bitumen were: penetration, softening point, flash point and water content.

2.1.1 Determination of Penetration

Specification test to measure the hardness of bitumen under specified conditions, in which the indentation of a bitumen in tenths of a millimeter at 25 °C (77 °F) is measured using a standard needle with a loading of 100 g and 5 s duration. It should be performed at least three determinations at different points of the surface [10].

2.1.2 Determination of Softening Point

Specification test in which the temperature is measured at which a bitumen (in the form of a disc under given loading conditions) softens and extends a fixed distance (25.4 mm) [11].

2.1.3 Determination of Flash Point

The flash point test determines the temperature to which an asphalt can be safely heated in the presence of an open flame. The test is performed by heating an asphalt sample in an open cup at a specified rate and determining the temperature at which a small flame passing over the surface of the cup will cause the vapors from the asphalt sample temporarily to ignite or flash [12].

2.1.4 Determination of the Water Content of the Product

Indicates the water content in the bitumen, since the presence of water in heating can cause explosions. This method is based on the fractional distillation of the bitumen whose constituents are separated due to the difference in boiling points of the liquids and the immiscibility [13].

2.2 Preparation of the Cement Pastes and Mortars

It was followed a 23 factorial design with replicate to prepare the experimental work. The variables were: Formulation: pastes (-) (Portland CPV ARI cement and water) and mortar (+) (Portland CPV ARI cement, water and sand), absence (-) or presence (+) of bentonite and with (+) or without (-) chemical additive (plasticizer).

The relationship between the amount of water and cement used for the paste and mortar were 0.35 and 0.58, respectively, and the ratio of the components of mortar was 1:2. The quantity of plasticizer and clay (bentonite) used was equivalent from 0.6% to 5% by weight of cement, respectively (Fig. 2).

2.3 Evaluation of Pastes and Mortars

The tests performed were determination of viscosity, setting time and compressive strength.

2.3.1 Determination of Viscosity

Viscosity is one of the properties in the cement paste which is a measure of the workability and determines the ease and homogeneity with which materials may be mixed, thus guaranteeing best solidified products. The optimum viscosity will depend on the mixing system used by the nuclear power plant [5].

2.3.2 Determination of Setting Time

It is the moment from which the water present in

the waste comes into contact with the cement, and begins the chemical reactions, whose consequence is gradual solidification. In practice, the setting time is an indication of compatibility between the waste and cement. It is an important property during the processing of waste, as it must be ensured that there is no solidification before the mixture was completely homogenized and it becomes a solid after the processing in order to be handled, stored and transported [14].

2.3.3 Determination of Compressive Strength

This test consists of subjecting the sample to the growing tension, continuing until its deformation or break, it is one of the most important mechanical characteristics. It is related to the degree of compression and the stiffness of the product. Therefore, products that are less minimum porous and more homogeneous, cracks and fissures tend to present more resistance to compression. The product must be resistant to impacts that may occur, especially during handling and transport [15].

2.3.4 Determination of Density

The density test was done by directly relate to the strength of the product. The denser the sample the lower is the amount of pores, leading to a higher resistance. For the determination of density was weighed the sample in an analytical scale and divided by the value obtained the same volume [5].

Fig. 2 Preparation of the cement pastes and mortars.

Fig. 3 The leaching test.

2.3.5 Determination of Leaching

The leaching test was based on the leaching of ISO 6961 by subjecting the samples to a lixiviant solution, followed by quantitative analysis of the chemical element (cesium) leached. The samples the leaching are sent to atomic absorption test to determine the amount of cesium leached (Fig. 3). The leaching test has a duration of 1 year [16].

3. Procedure for Selecting the Best Model

The results obtained in the tests for the bitumen used in Angra 2 are shown in Table 1.

The bitumen used in the immobilization of the waste of low and intermediate level of radiation Angra

2, is within the standard DIN 1995 [17].

The results from the tests with paste and mortar are presented in Table 2. For formulation B (mortar) not performed tests viscosity and setting time, because these tests are performed in LABCIM only pastes. The viscosity tests are performed only in the paste (formulation A) and it was not possible to do it in the paste without plasticizer and bentonite because it was very viscous.

The standard CNEN-NE-6: 09 establishes acceptance criteria for disposal of radioactive waste of low and intermediate level and specifies that the compressive strength at age of 28 days must be greater than or equal to 10 MPa [17]. All tests were within this specification. Then the formulation to be chosen has the best value and has the proper amount of bentonite, which will be important if it is necessary to absorb water or radionuclides.

It was observed that the use of Formulation A (paste) with bentonite and with plasticizer had higher values of compressive strength at age of 28 days and the lowest result was observed for formulation B (mortar) with plasticizer and with bentonite.

Table 1 Evaluation results of the bitumen.

Test	Results	Specification (DIN 1995)
Penetration (1/10 mm)	12.5	10-20
Softening point	70 °C	67-72 °C
Flash point	312 °C	>290 °C
% Water	< 0.1%	<1.0%

Table 2 Results in pastes and mortars evaluated.

Test	Formula-tion	Bento-nita	Plastici-zer	Density (g·cm ⁻³)	Viscosity (Pa.s)	setting time		Period	Compressive strength (MPa) (28 days)	Density 28 days (g·cm ⁻³)
						Start	End			
1	A	-	-	2,002	$6,765 \times 10^2$	01:24	02:44	01:20	30.4	1.98
2	A	-	+	2,010	$1,36 \times 10^1$	02:25	04:55	02:30	40.05	2.01
3	A	+	-	1,992	-	1:30	2:45	01:15	40.8	1.96
4	A	+	+	1,965	$1,148 \times 10^3$	1:31	2:37	01:06	41.2	1.96
5	B	-	-	2,100	-	-	-	-	28.2	2.08
6	B	-	+	2,192	-	-	-	-	29.2	2.19
7	B	+	-	2,135	-	-	-	-	24.1	2.11
8	B	+	+	2,085	-	-	-	-	23.9	2.04

Formulation A (pastes); Formulation B (mortar);
without bentonite (-); with bentonite (+);
without plasticizer (-); with plasticizer (+).

Fig. 4 Samples of cement pastes (left) and mortar (right) after being broken in the compressive strength test.

Table 3 Samples submitted to leaching.

Test	Formulation	Bentonita	Plasticizer
3	A	+	-
4	A	+	+
5	B	-	-
6	B	-	+

Formulation A (pastes); Formulation B (mortar);

without bentonite (-); with bentonite (+);

without plasticizer (-); with plasticizer (+).

The density values were higher for the mortar than for the pastes, though the latter ones have higher values of compressive strength at age of 28 days. This can be explained because it has sand that provides increase in the porosity of the specimen, resulting in reduction of resistance (Fig. 4).

For the realization of leaching test was selected the two best results the pastes and the mortar. The aim of this test is to assess the leaching, given that this is one of the basic parameters for the radioactive waste product disposal of according to CNEN-NN-6:09 standard. The Table 3 shows the leached samples, each containing 30 mg of cesium chloride [18].

The leaching test is in progress, but the first exchanges of leaching were carried out and the samples remained intact, not releasing any particles in leaching. Samples are being collected and sent for chemical analysis.

The first results are presented in Fig. 5 and it is observed that each exchange showed a peak in the graph due to accumulation of cesium leached during the exchange period [16].

5. Conclusion

The assessment of the bitumen used in the immobilization of the waste of low and intermediate level of radiation Angra 2, is within the standard DIN 1995.

For the evaluation of cement mortars and pastes the compressive strength at age of 28 days is considered a safety parameter relating to handling, transport and storage of the cemented waste product. The standard CNEN-NE-6.09 establishes that the compressive strength at age of 28 days must be greater than or equal to 10 MPa. All tests presented values above this specification. The best results of the compressive strength were obtained by using Formulation A (paste), with bentonite and plasticizer and the lower result was observed Formulation B (mortar) with plasticizer and bentonite.

For future work, will be finished the test of leaching in cement pastes and mortar and will be carried a new test of leaching using bitumen encapsulated on cement pastes and mortars that show better results in the leaching test.

Fig. 5 First leaching test results.

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