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STUDY OF BITUMINIZED WASTE PRODUCTS TO BE DISPOSED IN A NEAR-SURFACE REPOSITORY.

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ABSTRACT

The Project RBMN was launched in November 2008 and aims to establish, manage and execute all tasks for implementing the Brazilian Repository, from its conception to its construction. The concept to be adopted will be a near-surface repository. The inventory includes wastes from the operation of nuclear power plants, fuel cycle facilities and from the use of radionuclides in medicine, industry and activities research and development. The implementation of the national repository is an important technical requirement, and a legal requirement for the entry into operation of the nuclear power plant Angra 3. In Brazil, for the immobilization and solidification of radioactive waste of low and intermediate level of radiation from NPPs are used cement, in Angra 1, and bitumen, in Angra 2. Studies indicate serious concerns about the risks associated with bituminization radioactive waste, much related to the process as the product. There are two major problems due to the presence of products bituminization in repositories, swelling of the waste products and their degradation in the long term. To accommodate the swelling, filling the drums must be limited to 70 - 90% of its volume, which reduces the structural stability of the repository and the optimization of deposition. This study aims to evaluate of the properties of bitumen and cement pastes and mortars used in the immobilization of waste radioactive.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.

1. INTRODUCTION

In Brazil, there are currently two nuclear power plants with PWR type, located in Itaorna, Rio de Janeiro. The Angra 1 plant is in operation since 1983 and Angra 2, since 2000. They offer to the Brazilian electric power system 640 MW and 1,350 MW, respectively. In late 2009, the Federal Government announced the decision of the construction of Angra 3 Nuclear Power Plant, which is similar to Angra 2, expected to generate 1,405 MW [1].

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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 [2 ,3]. 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 radioactive waste. The responsibility of CNEN for the final destination of the waste is established in the Article 2. In accordance of this Law is not allowed receiving radioactive waste in liquid or gaseous forms in final repositories. 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 [4].

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The immobilization 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 a planned to be the bitumization used in a new nuclear power plant in Argentina. However, these countries abandoned and replaced by other processes, after the evaluation of the total cycle of waste from its generation to its disposal [6].

Experiences of bituminized waste disposal on surface repository were reported in countries like Romania, where ion exchange resins are immobilized in bitumen in 60 liter drums, which are then packed in drums 200L with an intermediate concrete shielding. Slovakia radioactive waste are incorporated into bitumen and this mixture is stored in drums 200L coated with zinc (galvanizing), deposited in containers and transported to the surface Mochovce repository. In the Czech Republic 100L waste drums are immobilized by high strength concrete in 200L drums, plates and galvanized corrosion-resistant paint, and then sent to the repository Dukovany [6].

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. The proposal of the Nuclear Energy Agency (OECD / NEA) 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 [7].

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 caused by fissures, softening by temperature variations, creep because it viscoplastic material, attack 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 an increases the area necessary for disposal [6].

The "RBMN Project" was launched in November 2008 and aims 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 [2].

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. EXPERIMENTAL

Initially it was made a literature review about the bituminization process and international experiences in the disposal of these products. Also researched the properties of bitumen used in Angra 2 for solidification of radioactive wastes.

The tests to evaluated the cement pastes (cement and water) and mortars (cement, water and sand) were organized using the 2^3 factorial design, in duplicate. The selected ones will be candidates to be used in the immobilization of the bituminized wastes.

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 5s duration (Figure 1). It should be performed at least three determinations at different points of the surface [8].

Figure 1. Equipment for determining the penetration.

2.1.2. Determination of softening point

Specification test in which the temperature is measured in °C at which a bitumen (in the form of a disc under given loading conditions) softens and extends a fixed distance (25.4 mm) (Figure 2) [9].

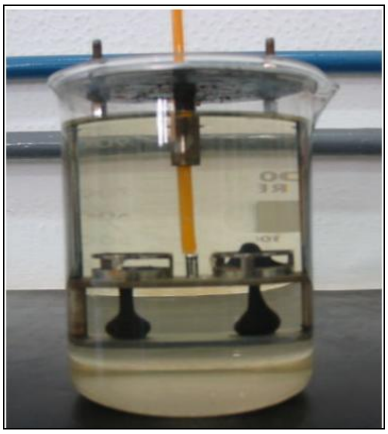

Figure 2. Equipment for the determination of softening point.

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 (Figure 3) [10].

Figure 3. Equipment for determination of flashpoint

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 [11].

2.2. Preparation of the Cement Pastes and Mortars

It was followed a 2^3 factorial design with replicate to prepare the experimental work. The variables were: Formulation: pastes (-) (cement and water) and mortar (+) (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.

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 [4].

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 [12].

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 [13].

3. RESULTS

The results obtained in the tests for the bitumen used in Angra 2 (VIT 70) are show in Table 1.

Table 1. Evaluation results of the bitumen.

Test	Results
Penetration	(11,5; 12,5; 13,0; 12,5; 12,5; 12,5) 1/10 mm
Softening point	70ºC
Flash point	312ºC
% Water	< 0,1%

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.

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

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 [14]. 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.

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

4. CONCLUSIONS

The assessment of the bitumen (VIT 70) used in the immobilization of the waste of low and intermediate level of radiation Angra 2, is within the manufacturer's specifications.

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 works, test will be performed in full-scale with the best formulation obtained in laboratory and then use this material to immobilize the bituminized wastes to evaluate this application.

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