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MICROWAVE PROCESS EMPLOYED TO STUDY THE IMMOBILIZATION FEASIBILITY OF SPENT ION EXCHANGE RESINS IN POLYMERIC MATRICES

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ABSTRACT

Nuclear activities generate radioactive wastes in several physical states, radioactive levels and kinds of radioactive emission. Hence, a lot of techniques have been developed and optimized to do the immobilization of these materials, according to local and international regulations to protect human being and environment. Another great concern is the indiscriminate disposal of used polymeric materials (such as plastic and rubber) or production leftovers in landfills, which remain for many years before they are naturally decomposed. In this work, it was studied the possibility of carrying out the immobilization of spent ion exchange resins (contaminated with ionising radiation), by using polymeric matrices of bitumen and rubber (as solidification materials for the storage of low level radioactive waste). The samples were mixed at different percentages and were heated in a microwave device (2,450MHz) at IPEN/CNEN-SP, varying the irradiation time and power. The objective of the immobilization is converting the wastes into forms that are leach resistant and physically and chemically stable for disposal. Characterizations of these materials have been performed according to ABNT-NBR standards. The results indicated the previous idea of the necessary minimum temperature to keep the matrix for future embedding of radioactive waste, in solid state.

1. INTRODUCTION

All nuclear activities, from the mineral extraction, physical and chemical changes to the final product, either in research, industry or medicine, generate radioactive wastes of varied physical states, radioactive levels and kinds of radioactive emission. These wastes may be solid, liquid or gas, with high, medium or low level of radiation and may also be alpha, beta or gamma emitters.

A lot of processes have been continuously developed and techniques have been optimized to improve the immobilization conditions of these wastes whose management must respect local and international regulations and laws that must show clear objectives to protect human being and environment both for our generation and the future of mankind.

Another great problem which is a consequence of fast increase of world's population regards the indiscriminate disposal of polymeric material leftovers, such as plastic auto parts, toys, shoe soles, tires, hospital materials, packages among others in landfill where they remain for many years before they are naturally decomposed.

This work contemplates the development of immobilization technology by using the heat of a microwave device (2,450 MHz) at IPEN-CNEN/SP, varying the irradiation time and power, to carry out the immobilization of spent ion exchange resins by using polymeric matrices of bitumen and rubber from production leftovers (Ethylene Vinyl Acetate – EVA, silicone and neoprene®).

One of the polymeric materials mostly used in the immobilization of radioactive wastes is bitumen because it allows materials of different physical and chemical characteristics to be incorporated in the same matrix. Bitumen is a generic term that covers a wide range of high molecular hydrocarbons [1]. Its main characteristics that make it suitable as a matrix are insolubility in water, high resistance to water diffusion, plasticity with aggregate, stable blend, good rheological properties and easy application. These are important characteristics when related to activities of transference and storage of melted bitumen, to operational conditions to be compatible with the wastes and to packaging requirements and final disposal [2,3].

The most important characteristics of microwave technology are the rapid and selective heating and the environmentally clean process with low pollutant emission. The microwave heating occurs when the electromagnetic waves penetrate the material, usually polar molecules, and release energy as heat [4].

2. EXPERIMENTAL

2.1. Samples Preparation

Two groups of samples lots were prepared. In the first group (Table 1) it was analyzed the increment of mechanical resistance to matrix of pure bitumen caused by addition of other polymers (0% to 15% in weight).

Table 1. Samples composition (% wt) with bitumen and rubbers.

Samples composition	**100% A**	**95% A 5% B**	**95% A 5% C**	**95% A 5% D**	**90% A 10% B**	**90% A 10% C**	**90% A 10% D**	**85% A 15% B**	**85% A 15% C**	**85% A 15% D**

A – Bitumen; B – EVA; C – Silicone; D – Neoprene®.

The second group (Table 2) was compounded by bitumen and the rubber that showed the best performance in the first group, EVA rubber. In that matrix the ion exchange resin was immobilized.

Table 2. Samples composition (% wt) with bitumen, resin and EVA.

Sample	**11**	**12**	**13**	**14**	**15**	**16**	**17**	**18**	**19**
Bitumen (%)	50.0	47.5	45.0	47.5	47.5	47.5	45.0	42.5	40.0
Resin (%)	50.0	52.5	55.0	47.5	42.7	37.5	40.0	42.5	45.0
EVA (%)	0.0	0.0	0.0	5.0	10.0	15.0	15.0	15.0	15.0

2.2. Samples Irradiation

All samples with 400g of total weight were separated and irradiated inside a glass vessel, by using electromagnetic waves in a microwave device (power supply of 1,000W and frequency of 2,450MHz) at IPEN-CNEN/SP. This equipment consists of a magnetron valve, a wave-guide and a cavity where the samples were irradiated. The effect of the following process parameters was evaluated: irradiation time and microwave power supply. The temperature was controlled by a "K" thermocouple.

2.3. Samples Characterization

All the samples of the first group were submitted to the following tests: penetration, softening, flash and fire points, to identify the matrix that showed the highest mechanical resistance results. The lot of samples of the second group was submitted only to the penetration test to identify the matrix that presented the best immobilization efficiency and mechanical resistance.

Tests description

- Penetration test: the Brazilian standard NBR 6576 (1998) covers determination of penetration of semi-solid and solid bituminous materials. Penetration is the distance in millimeters that a standard needle penetrates the sample vertically, under predetermined load conditions with 100g of sample in 5s at 25°C.
- Softening point test: the Brazilian standard NBR 6560 (2000) covers the determination of the softening point of bitumen. The ring and ball apparatus is used to determine the transition point between solid and liquid state and to indicate the trend of the material flow under that temperature.
- Flash and fire points test: the Brazilian standard NBR 11341 (2004) describes the determination of flash and fire points of petroleum products by a manual or an automated Cleveland Open Cup apparatus.

3. RESULTS AND DISCUSSION

The matrix behavior, varying the percentage of EVA, obtained in the first group is shown in Figures 1 and 2. The addition of EVA led the softening point to reach 120°C while the softening points of the other rubbers were kept between 85°C and 95°C.

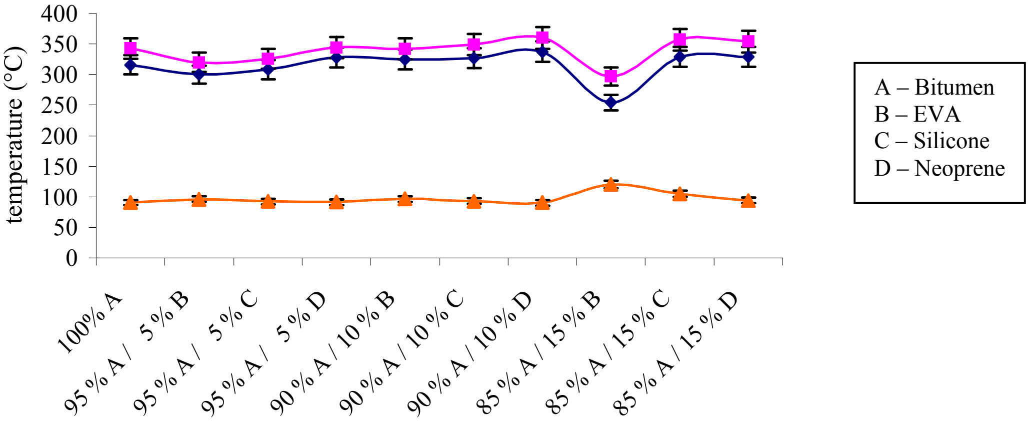

Figure 1. Results of fire (pink line), flash (blue line) and softening (yellow line) tests as a function of sample compositions.

From these results, the temperature monitoring was done along with the process to prevent the bitumen and resin temperature from reaching 250°C (flash point) and 160°C (releasing explosive amino acids), respectively.

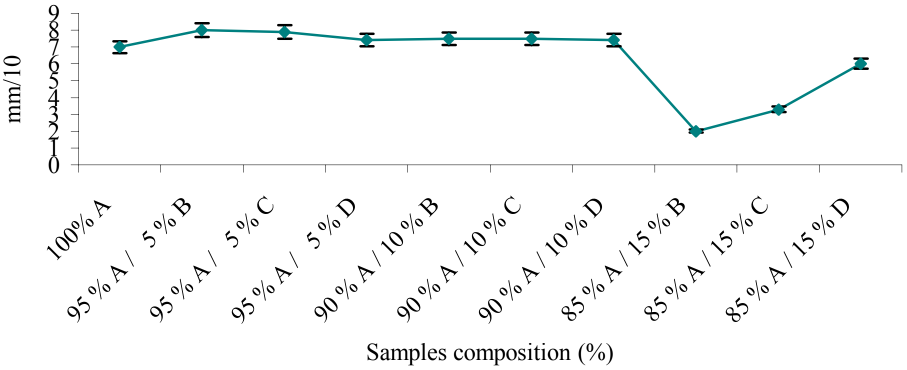

Figure 2. Penetration test of samples from group 1.

Based on the sample that reached the best mechanical resistance or the least value of penetration test (sample composition – 85%A/15%B), a new group of samples was prepared with addition of ion exchange resins in the amounts indicated in Table 1.

The values of penetration test for the second group are shown in Figure 3 where it can be seen that the resistance remained stable (as it can be seen in the linear portion in Figure 2) with the addition of the resin to the matrix bitumen/EVA. These data indicate a previous measure of material deformation. The smaller was the penetration measure the higher was the matrix hardness, and the compression resistance increased when it was compared to a matrix consisted of pure bitumen. Regarding silicone rubber and Neoprene®, the results did not show significant changes when compared to the achieved data of pure bitumen.

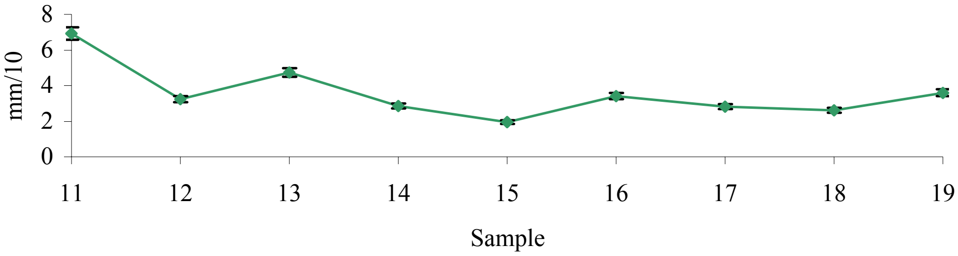

Figure 3. Penetration test of samples from group 2.

Concerning those results, it was possible to get a previous idea of the necessary minimum temperature (up to120°C) to keep the matrix in solid state for future embedding of radioactive waste.

A series of assays to determine the irradiation time and the microwave power applied in the samples was performed for pure bitumen samples. Fifteen minutes of irradiation were necessary by keeping the microwave power applied in 100%.

Also a similar series of assays was performed for polymer, and within five minutes of irradiation under 100% of microwave power applied, the ideal temperature was reached by addition and homogenization of the blend.

The choice of both matrices with incorporated resin can be refined through additional chemical tests. These tests will give evidence of leaching resistance of the matrices.

Concerning the radiation level, the blend stability will be studied to define the maximum amount of waste that could be incorporated in that matrix.

3. CONCLUSIONS

It is important to point out that the microwave irradiation time and power applied to optimize the process were controlled to keep the safety limits established for those materials (bitumen, EVA, silicone, neoprene), providing fast, uniform and selective bulk heating at them.

The efficiency of microwave technology for immobilization of radioactive waste has been evaluated with good results but its application on this area is still in the first steps of development.

Based upon the characterization results it was possible to verify that the addition of rubber in the bituminous matrix improved the performance regarding the mechanical properties, which kept their characteristics after the addition of 47%wt of resin (Sample 15) with the least value of penetration test.

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