PREPARATION OF POLYMERIC MATRICES BY USING MICROWAVE ENERGY FOR IMMOBILIZATION OF RADIOACTIVE WASTE

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Abstract - The management of radioactive hazardous is not simple and many treatment processes have been studied to ensure that the material is handled safely. Thus the continuous improvement of waste immobilization conditions must always be considered. Also, there is another problem concerning the discharge of leftovers of polymers production in landfill, where they remain for many years, before they are naturally decomposed. In this work, the possibility of reusing of vulcanized rubber (polymeric material) was analyzed as a component of bitumen matrices for immobilization of radioactive waste. The bituminization process has been widely used to embed low and intermediate level radioactive waste. The preparation of matrices and generation of homogeneous blends of those materials were made by using microwave radiation, as heating supply that occurs when the electromagnetic waves penetrate the material, usually polar molecules, and release energy as heat. According to the preliminary results of the studies was obtained the better ratio of bitumen/polymer (EVA), suitable for incorporating radioactive waste (ion exchange resin), considering the samples characterization by using tests of penetration, softening point, flash and fire points.

Introduction

The cycle of development, manufacturing and use of nuclear materials has aroused a serious matter: the production of radioactive waste, whose management is not simple and several treatment processes have been studied to ensure that the material is handled safely.

Thus a matter must always be considered: the continuous improvement of waste immobilization conditions, in more stable form, which would be able to keep its insulation during its entire decay and to protect the human being and the environment from harmful radiation effects. The storage of those residues depends on the composition, activity level and disposal options for the final product. It also must be done by applying chemically stable materials as immobilization matrices in order to avoid as much leaching and dispersion of compounds as possible.

Nowadays, there is another problem concerning the discharge of leftovers of polymers production in landfill, where they remain for many years, before they are naturally decomposed. Those leftovers are usually the result of the automobile manufacturing plastic, toys, shoe soles, tires, hospital materials and so on [1].

In this work, the possibility of reusing polymeric materials was analyzed particularly, vulcanized rubber, as a component of bitumen matrices for immobilization of radioactive waste. The bitumen is a compound, known as asphalt, employed in paving and impermeabilization. It is the most important component of asphalt which is obtained from petroleum distillation. It is a semi-solid material with 85%wt up to 95%wt of carbon and, in small amounts, hydrogen, sulphur, nitrogen and oxygen. The bitumen has some advantages like: plasticity with the aggregate, impermeability, stable blend and easy application. In this study, the material was used as agglomerate agent in the immobilization matrices of radioactive waste, helping the reduction of leftovers and showing resistance to leaching, radiation and aging.

The bituminization process has been widely used to embed low and intermediate level radioactive waste. This method allows a stable physical, chemical and radiological board, in many levels of radioactive waste management [2]. In general, in these immobilization processes the mixture of polymeric matrix and radioactive waste are made in an extruder, where the materials are heated (although the equipment might be contaminated). Next, the mixture is poured in barrels for final storage where it becomes solid as it gets cold. This is the most usual method because it brings excellent results [3].

The preparation of matrices and generation of homogeneous blends of those materials were made by using microwave radiation, as heating supply. 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].

Therefore, this work aimed at showing the results of the studies that determine the ratio of bitumen/polymer (ethylene vinyl acetate - EVA), suitable for incorporating radioactive waste (ion exchange resin), in a stable matrix, by applying microwave energy as heating supply.

Experimental

2.1 Samples Preparation and Irradiations

The samples were prepared and irradiated by using glass ware in order to better define the proportion of materials that should be used in the immobilization of radioactive waste (contaminated ion exchange resin) with bitumen and rubber.

The percentage of bitumen in the blend resin/rubber (total weight of 400g) was: bitumen - 40.0%wt to 50.0%wt; resin - 45.0%wt to 50.0%wt and rubber - 0.0%wt to 15.0%wt. The involved material properties were: bitumen – asphalt VIT–90, Viapol; rubber - EVA (shoe sole leftover, Grendene®); inactive (non contaminated by radiation) ion exchange resin - Amberlite® – IRN 217 (type anionic-cationic, sphere diameter of 0,3mm-0,2%, formulated to nuclear application – primary circuit water chemical treatment in the research reactor) and magnetite powder Fe_3O_4 with density of 5,3g/cm3 at 25°C and used as microwave absorbing [5, 6, 7, 8].

The irradiations were carried out in a microwave device (microwave power supply 1,000W – Panasonic - frequency 2,450MHz), at IPEN-CNEN/SP. This equipment consists on 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 (1min up to 15min) and microwave power supply (5% up to 100%). The samples temperature was controlled by using a “K” thermocouple.

First step: bitumen and magnetite irradiations for 15min (according to experimental arrangement in figure 1) and cooling time of 5min to distribute the temperature equally in the recipient.

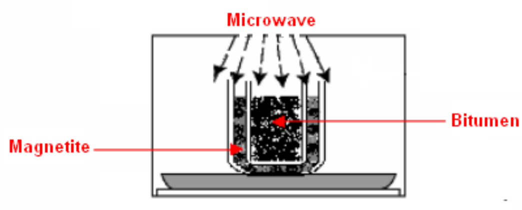

Figure 1 – Bitumen and magnetite irradiations in the microwave oven.

Second step: Blend of resin/rubber and magnetite irradiations for 5min (according to experimental arrangement in figure 2).

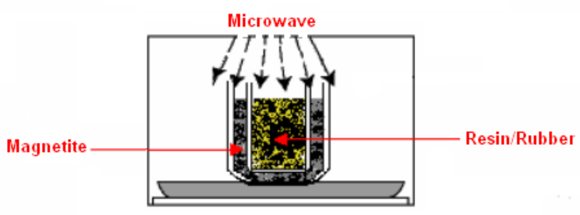

Figure 2 – Resin/rubber and magnetite irradiations in the microwave oven.

2.2 Samples Molding

All the blends of resin/rubber were put in paraffin paper cups and the bitumen was added. The mixture was manually shaken to obtain more homogeneity. After some cooling time, the molds were separated in samples (matrices) to be characterized.

2.3 Samples Characterization

The matrices obtained were analyzed by the tests of penetration, softening, flash and fire points to identify which of them could show the best immobilization efficiency and the highest mechanical resistance.

Penetration test: Brazilian standard NBR 6576 (1998) - penetration of semi-solid and solid bituminous materials. Penetration is the distance (in millimeters) that a standard needle penetrates the sample vertically, under a predetermined load conditions, with 100g of sample in 5s at 25oC.

Softening point test: Brazilian standard NBR 6560 (2000) - softening point of bitumen. The ring and ball apparatus is used to determine the transition point between solid and liquid state and indicate the trend of the material flow under that temperature.

Flash and fire points test: Brazilian standard NBR 11341 (2004) - flash and fire points of petroleum products are obtained by using a manual Cleveland open cup apparatus or an automated Cleveland open cup apparatus.

Results and Discussion

The bitumen temperature during the first step was not over 250°C (flash point temperature) and it was under 85°C (softening point), respectively. In the second step, this temperature was controlled between 100°C and 160°C to make possible mix rubber and resin without the bitumen become hard or solid.

According to the results in the flash point test, it was possible to determine the safe operation temperature of the materials employed in the matrices production. 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.

Several irradiations were performed to determine the irradiation time and the microwave power applied in the samples utilizing pure bitumen. It was possible to irradiate this sample for 15min, by keeping the microwave power applied in 100%, to get the necessary fluidity to prepare the matrices, without degrading the sample. Also it was observed that under 100% of microwave power applied, 5min of irradiation were necessary to reach the ideal temperature by addition and homogenization of the blend resin/rubber without exceeding 160oC. From these results (Table 1) it was noticed that the addition of EVA rubber in the immobilization matrix reduced the penetration value from 6.8 (S1 sample – 50.0%wt bitumen/50.0%wt resin/0.0%wt rubber) to 1.9 tenth part of millimeter (S5 sample – 47.5%wt bitumen/42.5%wt resin/10.0%wt rubber).

These data indicated 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.

Table 1 - Results of penetration test for blends (bitumen/resin/rubber).

Sample	Penetration (1/10mm)
S1	6.8
S2	3.4
S3	3.3
S4	2.9
S5	1.9
S6	2.7
S7	2.8
S8	2.6
S9	3.7

The matrix behavior, varying the percentage of EVA, can be seen in Table 2. The addition of EVA led the softening point to reach 120°C.

Table 2 - Softening point of bitumen/EVA matrix.

Sample	Temperature (°C)
100%/0.0%	90
95.0%/5.0%	95
90.0%/10.0%	105
85.0%/15.0%	120

Concerning these results, it was possible to get a previous idea of the necessary minimum room temperature to keep the matrix for future embedding of radioactive waste, in solid state.

Conclusions

The addition of polymer in the bituminous matrix improved the performance regarding the mechanical property. Herewith, it could be assumed an increase in the resistance to weariness, in the permanent deformation and a reduction of thermal crack for the developed blend, when it was compared with the matrix of pure bitumen. For mechanical resistance, EVA rubber showed the most suitable results compared to those obtained to pure bitumen.

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.

The efficiency of microwave technology for immobilization of radioactive waste has been evaluated and its application on this area is still in the first steps of development. In the present study the microwave irradiation time and the microwave power applied to optimize the process were controlled to keep the safety limits established for those materials.

The advantages of using microwave energy for processing hazardous wastes depend on the type and characteristics of the wastes to be treated. It can be cited some important features of this technique: rapid heating, high temperature capabilities, selective heating, ability to process wastes in-situ, treatment or immobilization of hazardous components to meet regulatory requirements for storage, transportation or disposal and energy savings. This system is particularly suited to treatment of hazardous or radioactive materials.

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