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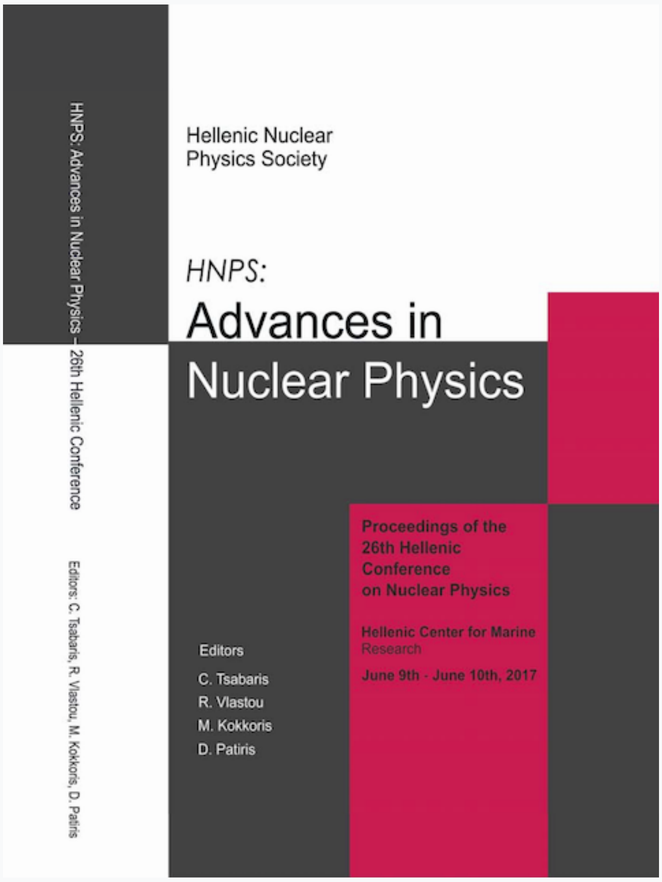

Technique for Characterization and Clearance of the Resin Waste from the Greek Research Reactor (GRR-1)

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Technique for Characterization and Clearance of the Resin Waste from the Greek Research Reactor (GRR-1)

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Abstract

The present work concerns the examination of management options for VLLW drums [1] with ion exchange resin waste from the Greek Research Reactor (GRR-1). According to the general clearance criterion [2, 3], after cementation, 75 % of the VLLW could be cleared and for the remaining 25 % the summation formula result is lower than 1.6. The proposed management option for the total amount of the VLLW resin is the spread of the cemented resin, before thickening, over the ground around the interim storage facilities, inside the controlled area, for pavement construction. Additionally, a quantity of the cemented waste will be used for preparation of blocks for quality and homogeneity examination of the waste form.

Keywords: resin waste; clearance; waste cementation

1. Introduction

The ion exchange resin waste, which belongs to the VLLW class, is kept in primary form at the old radioactive waste interim storage facility of the National Centre for Scientific Research "Demokritos" and constitutes the 60% of the total resin waste quantity.

The total amount of resin waste is about 16 Mg and arose during the GRR-1 operation period 1961-2002. Radioactivity levels were evaluated in each resin drum by sampling and analysis using gamma spectrometry. The determined activity concentrations are: Ag-108m < 4.70 Bq/g, Cs-137 < 160 Bq/g, Eu-152 < 2 Bq/g and Co-60 < 6 Bq/g.

In a previous study, it was shown that the total quantity of the resin waste could be cleared for disposal at a landfill within few decades [4]. Nevertheless, the landfill disposal option is not directly applicable in Greece, since there is no such installation and neither a plan for construction. For this reason, another management option for the resin waste which belongs to the VLLW class was necessary to be explored in order to make space inside the storage facility.

It is considered that a mixture of 10% of resin and 90% of cement is necessary to satisfy the requirements of stability for a long-term storage [5, 6] and furthermore to prevent dispersion of radionuclides and other pollutants into the environment.

2. Ion exchange resin of the GRR-1

The resin waste was produced from the demineralization co-flow regenerated units of the GRR-1. The total amount is 16 Mg and is separated into 158 drums of 100 kg each. This waste arose during the reactor operation period 1961-2002 and is kept in

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primary form at the interim storage facilities for radioactive waste at the National Centre for Scientific Research "Demokritos".

GRR-1 is an open pool-type research reactor. It went critical for first time on June 1961. Since April 1964, the reactor operated at thermal power 1 MW. In 1971 the reactor was upgraded to 5 MW with change of the fuel elements from Low Enriched Uranium (LEU) to High Enriched Uranium (HEU). In 1990 a further upgrade of the reactor was performed by the introduction of beryllium reflector blocks. In the time period from 1999 to 2004, gradual replacement of spent HEU fuel elements by LEU elements took place. The reactor stopped operation in 2004 and now it is in extended shut down.

The ion exchange resin is a styrene divinylbenzene copolymer of density 0.84 g cm^{-3}. It was used for purification of the reactor coolant water at the demineralization co-flow regenerated units. After the end of its useful life, the resin material of total mass 300 kg was stored for one year for radioactivity decay at the reactor premises and then was put in 3 steel drums of 200 l, internally coated with plastic. Each drum was labeled as regards the year of receipt by the radioactive waste management section. The storage period of resin, until 2016 (the year of decay correction), ranged between 14 to 54 years, thus the shorter lived radionuclides have been decayed.

3. Radiological Characterization of the resin waste

The radiological characterization of the resin waste was carried out by collecting samples of primary waste from each of the resin drum and performing analysis by gamma spectrometry. The analysis results indicated that the waste belongs to the classes VLLW and LLW [1]. The activity concentrations were: Ag-108m < 4.70 Bq/g, Cs-137 < 160 Bq/g, Eu-152 < 2 Bq/g, Co-60 < 6 Bq/g. The dose rate on the surface of the drums ranged between background and 4 μSv/h.

The 95 drums with VLLW resin waste are kept at the old radioactive waste interim storage facility. It should be mentioned that VLLW contains radionuclides with activity concentration in the region of or slightly above the levels specified for the clearance of material from regulatory control without any restriction [2, 3].

The characterization of the resin waste was performed during the time span from 2003 to 2006. Two (in some cases three) samples of about 200 g resin were collected from different depths of each waste drum. The typical procedure for activity determination required measurement of each sample using a NaI(Tl) detector gamma spectrometry system. The detector system consisted of a Bicron Monoline scintillation detector NaI(Tl) (Model 3M3/3), the lead shielding structure with thickness 5 cm and a PC-based digital signal processing unit.

Analysis was performed for determination of Ag-108m, Cs-137 and Co-60 isotopes using the 433.9 keV, 662 keV and 1332.5 keV gamma ray emission lines, respectively. The 433.9 keV gamma ray emission line of Ag-108m was chosen in order to avoid possible interference from the Cs-137 line at 662 keV while the 1332.5 keV line of Co-60 was selected since the gamma ray background counts are the minimum at this energy region. These isotopes are the main presented in the resin waste. In some samples Eu-152 was detected and it was determined using the 340 keV gamma ray emission line. Since the minimum detectable activity (MDA) of Eu-152 was not low enough in most of the cases (higher than the one tenth of the general clearance limit of Eu-152 that is 0.1 Bq/g), the MDA of Eu-152 that was determined from each specific spectrum was used besides the determined activities of the other three radionuclides in the summation formulas for clearance. Nevertheless, representative samples were transferred to a HPGe based high resolution gamma spectroscopy system for peak identification and analysis in the framework of an inter-calibration between the two spectrometry systems [7]. The duplicate measurements for each drum were averaged in order to determine the radioactivity levels.

Furthermore, the data of gross beta and gamma spectroscopy analyses of liquid wastes that were being produced during the regeneration procedure were used to confirm that the radionuclides which should be taken into account in the summation formula for verification of clearance are the determined gamma emitters [4].

4. Clearance calculations

The characterization results were used in order to perform the clearance calculations, based on the summation formula (1):

$$\sum_{i=1}^{n} \frac{C_i}{C_{Li}} \leq 1 \quad (1) \quad \text{where,}$$

C_i the total activity in the structure per unit mass of the radionuclide i (Bq/g),
C_{Li} the clearance level of the radionuclide i (Bq/g),
n is the number of radionuclides in the mixture.

In the above formula, the ratio of the concentration of each radionuclide to the clearance level is summed over all radionuclides in the mixture. If this sum is less than one, the material complies with the clearance requirements.

5. Classification of resin waste

The activities of the detected radionuclides Ag-108m, Cs-137, Co-60 and Eu-152 in the primary resin waste that were calculated during the time span 2003 to 2006, corrected for 01/12/2016. The mean radioactivity concentrations were estimated for each of the 95 VLLW drums. The clearance levels which are mentioned in the Greek Regulation [2] for Eu-152, Ag-108m and Co-60 is 0.1 Bq/g and for Cs-137 is 1 Bq/g. The general clearance limits referred in the Greek Regulation are in accordance with the EU publication RP 122 (Part I) [3].

The clearance calculations showed that 12 drums of the primary resin waste satisfy the general clearance criterion and could be removed from the regulatory control. After cementation, 63 drums could be cleared and 20 drums (the summation formula result is lower than 1.6) could not, according to the general clearance criterion (Table 1).

It is considered that a mixture of 10% of resins and 90% of cement is necessary to satisfy the requirements of stability for a long-term storage as well as to prevent dispersion of radionuclides and other pollutants into the environment [5, 6].

Table 1

Clearance calculations results for the 95 VLLW drums

Summation Formula range	Number of resin waste drums in primary form	
0.01 - 1.00	12	Drums that could be cleared
1.01 - 2.80	18	Drums that may satisfy the general clearance criterion after cementation
2.81 - 4.60	17	
4.61 - 6.40	13	
6.41 - 8.20	9	
8.21 - 10.00	6	
10.01 - 11.00	3	Drums that could not satisfy the general clearance criterion after cementation
11.01 - 12.00	8	
12.01 - 13.00	2	
13.01 - 14.00	2	
14.01 - 15.00	2	
15.01 - 16.00	3	

6. Proposed management option for the resin waste

After performing specific measurements for clearance verification (see §7) the following management option is proposed:

- The 12 drums with primary resin waste which satisfy the general clearance criterion can be removed from the regulatory control and solidified in cement for strength testing, manufacture of cement shielding blocks etc.
- The 63 drums, which after cementation of the resin waste may satisfy the general clearance criterion, can be also released. A practical solution could be the spread of the cement before thickening over the ground around the interim storage facilities in order to prepare the lower and the upper layer of a pavement. Iron grid will be used and gravel will be added in the cemented resin mixture [5, 6].
- The 20 drums having result of the summation formula, after cementation, between 1.0 and 1.6, despite the fact that they do not satisfy the general clearance criterion, they could be used for preparation of the intermediate layer of the pavement.

7. Clearance verification procedure for the resin waste

The procedure which will be followed for clearance verification of each primary resin waste drum is:

- The drum will be measured externally at an area of low background by a non-destructive gamma spectrometry technique based on NaI detector [7].
- The specific mass activities will be determined for the primary resin waste.
- The determined specific activities will be used in the summation formula to confirm the preliminary evaluation based on sampling.
- The drum will be opened and the surface of primary resin will be checked with a contamination monitor for gross beta emitters for reconfirmation [4].
- In case the gross beta cps is comparable with total beta gamma cps, the procedure of cementation of resin waste will be carried out.
- The ratio of the ingredients for the production of the cemented resin mixture will be maintained (see §8).

8. Examination of homogeneity and activity determination of the cemented waste

Before starting the cementation of the resin waste, the procedure of cementation will be examined in regards with strength and homogeneity of the produced mixture.

For strength testing, primary resin waste from the 12 drums, which satisfy the general clearance criterion, will be solidified in cement for construction of blocks.

For homogeneity testing, blocks with dimensions 30cm x 20cm x 10cm of cemented resin waste with the highest concentrations in Ag-108m and Co-60 will be produced. These blocks will be monitored by a gamma spectrometry non-destructive technique in order to compare the different spectral data. The analysis will be performed for these isotopes which are considered typical for the waste material. The specific activity in the primary resin waste for Ag-108m in the drum #91 is 1.2 Bq/g and for Co-60 in the drum #149 is 0.7 Bq/g. These blocks, after monitoring, can be also used for shielding purposes in the radioactive waste interim storages.

It should be mentioned that the homogeneity of cemented resin waste depends on factors such as the operation procedure of the cement mixer and the water percentage of the mixture. As a result, these factors should be altered accordingly in order to ensure the homogeneity of the cement-resin waste mixture. When the homogeneity is achieved, the blocks will be measured by a gamma spectrometry non-destructive technique. Then, the specific activities will be estimated in order to determine the percentage of resin in the mixture. The factors of the cementation procedure which provide homogeneity and robustness will be maintained during preparation of the cement-resin mixtures.

A non-destructive technique for the blocks geometry will be developed by the use of MCNP code. Currently, a preliminary evaluation was carried out. The detector was simulated as NaI cylinder with dimensions 3" x 3" and the source as 30cm x 20cm x

10cm parallelepiped of common cement. The main axis of symmetry of the parallelepiped was on the NaI cylinder axis and its center at 19.2 cm from the surface of the NaI cylinder axis. Furthermore, the detector efficiency for point sources placed on the detector axis at 19.2 cm from the detector surface was evaluated by a MCNP model which was validated by the use of four gamma standard point sources Co-60, Ba-133, Cs-137 and Am-241. The evaluated detector efficiencies for point sources and volume source (common cement block) by MCNP, as well as the experimental data for the point sources are presented as a function of the peak energy in Fig. 1.

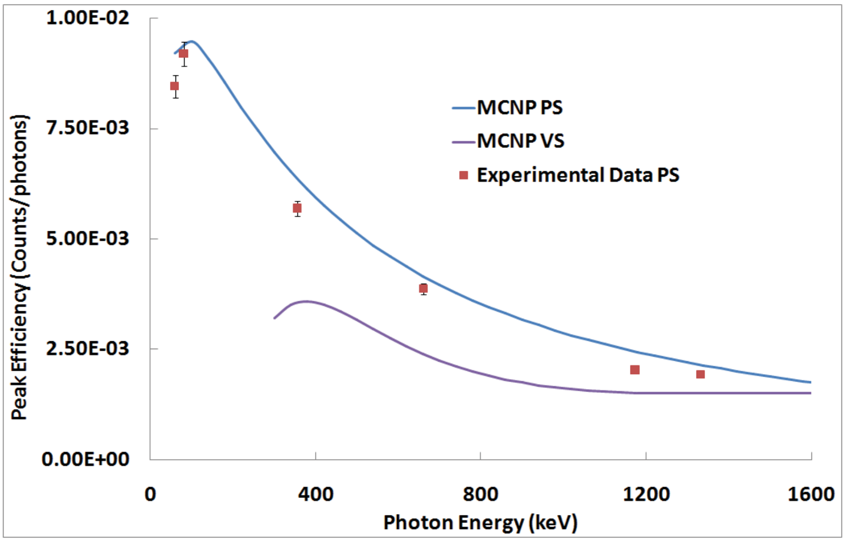

Fig. 1. Evaluated detector efficiencies for point (PS) and volume (VS) sources and experimental data for point sources (PS)

It is obvious from Fig. 1 that the evaluated efficiency for the cement block (VS) seems to be adequate for development of a sensitive measurement technique. Moreover, despite the bias, the experimental data for point sources are in good agreement with the evaluated data.

9. Conclusion

Resin waste classified as VLLW can be disposed at a landfill or a near surface disposal facility. These solutions are not directly applicable in Greece. For this reason, the solution of construction of a pavement with the cemented resin waste around the radioactive waste interim storage facilities, inside the controlled area, is proposed. By this way, 95 drums with resin waste could be cleared and removed from the interim storage facility of NCSR "Demokritos".

The proposed procedure and the development of a non-destructive technique for homogeneity testing seem feasible and can be implemented in case of approval by the Greek Atomic Energy Commission (EEAE).

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