DETERMINATION OF DIFFICULT TO MEASURE NUCLIDES IN SPENT RESINS

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This paper presents a method to determine Strontium – 90 in liquid samples by extracting Strontium 90 from the sample with the Eichrom resin, Sr-Spec, and using the Deconvolution Method in order to eliminate the interferrences due to other radionuclides. Due to its pure beta emission, Strontium 90 cannot be measured directlz bz radiometric methods, and it requires radiochemical separation from the matrix and determination bz liquid scintillation counting afterwards. The experiments were performed at the Nuclear Research Institute, Piteşti, Romania and the method provided by Eichrom to extract Stronntium 90 from the sample was adapted to the laboratory's resources.

Keywords: Strontium 90, deconvolution, radioactive waste.

1. Introduction

The Radiological characterization represents one of the basic processes in radioactive waste management activities, through which it can be achieved the goal of a high quality final product realization for disposal. In order to properly characterize the radioactive waste packages it is required to know the amounts and concentration of specific radionuclides in the waste package. Many of these specific radionuclides are difficult to measure from outside of the package, as they are alpha and beta emitting radionuclides, and they require laborious and complex radiochemical separation methods, which are not practical for large amounts of waste packages originating from the same waste stream [1].

The difficult-to-measure nuclides of primary interest are those with very long half-lives which will persist in a disposal site long after the period of institutional control. Their declaration is often important for the assessment of the consequences to health and safety for future uses of the disposal site. The information about the activity concentration and total activity are also required for the transport of radioactive materials [2].

90Sr can be produced by neutron fission in nuclear reactor and weapons testing and released to environment and it is one of important radionuclides in the views of radiation protection, environmental monitoring, radioecology, and radioactive waste management due to its relative high radioactive level in

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environmental and nuclear waste samples [3]. It has a half life of 29.1 years and maximum beta emission energy of 546 keV.

This paper aims to present a method proposed for the content of Strontium 90 determination, which is a difficult to measure nuclide present in almost all radioactive waste packages whose waste stream origin is the spent fuel, as fission product. This power plant has heavy water as coolant and moderator. Moreover, the moderator and the coolant are separated and form two different systems. The determinations and analysis of the resulted data has been performed at the Research Institute for Nuclear Reactors, Piteşti, Romania.

In the spent resins from a nuclear power plant, Strontium 90 originates from the fuel. In case of a fuel failure it will enter the cooling agent and then the purification systems with ion exchange colums. The ion exchange columns have the role to retain activated corrosion products and some fission products and minor actinides, and Strontium 90 is one of these. The first objective when we want to determine difficult to measure nuclides in radioactive waste samples is to dissolute the sample. In the case of spent resins this step is a very problematic one due to the nature of this type of sample and the dissolution can be made by dissolution in special reagents (e.g. Fenton's reagent) or by using a special equipment. After the dissolution is complete the sample is ready for Strontium 90 separation.

2. Experimental

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The method proposed in this paper for the determination of 90Sr is by using a Sr-Spec resin and a Liquid Scintillation Counter. The experiments were performed using the procedure provided by Eichrom for the determination of strontium in water and adapting it to the laboratory's resources and the main objective of this experiment was to validate this method and to use it in the future for the determination of Strontium 90 not only in spent resins, but also in other types of wastes, such as effluents.

The experiment was performed in two runs in parallel. Two initial samples were prepared and both consisted in a mixture of 1 ml sol 90(Sr+Y) in equilibrium in 0.1M HNO3 with a concentration of 258 Bq/g ± 5% in 01.08.2006 and 1 ml Sr tracer with a concentration of 5 mg/ml (the Strontium tracer was used in determining the gravimetric yield). Using two columns (one column for each sample) with Sr Resin provided by Eichrom, Strontium 90 was separated from Yttrium 90. The extractant system in Eichrom's Sr Resin is 1.0M 4,4' (5') - di-t-butylcyclohexano 18-crown-6 (crown ether) in 1-octanol [4]. The solutions were evaporated and the residue was dissolved in a small quantity of 8M HNO3 solution.

The resin was placed in two 5ml plastic columns and conditioned with 10 ml 8M HNO3. After conditioning the initial solutions were allowed to be drained through the resin columns. The resin has the property to adsorb strontium and let the others elements pass through the column. The resulting solutions which are called the non-adsorbing parts are labelled in the next steps Y1 and Y2. Next the adsorbed strontium was eluted with 10 ml 0.05M HNO3. The resulting solutions are Sr1 and Sr2. To calculate the recovery the eluate is evaporated to dryness, and then the net mass of residue is measured. Strontium salt was redissolved with 1M HNO_3 and analyzed by Liquid Scintillation Counting (Packard TriCarb 2100TR). The scintillator used was UltimaGold AB™ which has a good capacity for acid solutions. Fig. 1 shows the diagram of Strontium 90 separation.

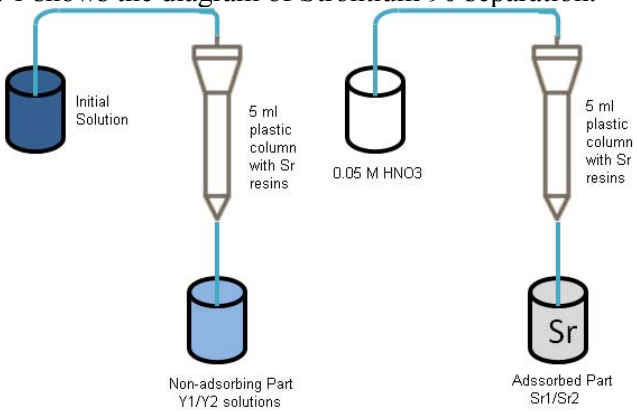

Fig. 1. Diagram for the Strontium separation

3. Results

After extraction, the samples have been prepared for the Liquid Scintillation Counting Analysis. The analysis was performed the next day and due to the desintegration of Strontium 90 to Yttrium 90 we could observe the Yttrium ingrowth in the sample. Figure 2 shows the spectra obtained for the non adsorbed

part solutions. As it can be seen both Y1 and Y2 contained only Ytrium, which proves the efficiency of the resin regarding Strontium adsorbtion.

Table 1 presents the activity, in Bq, of the analyzed solutions. The recoveries of Ytrium in the non-adsorbed solutions is 95% and 98%.

Table 1

Activity of Y1 and Y2 solutions

Sampe No.	Initial sln.	Y1	Y2
Activity [Bq]	129	123	127
Recovery [%]	-	95	98

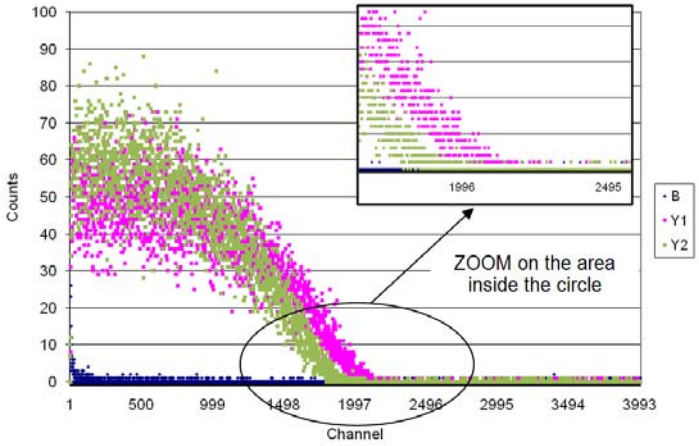

Fig. 2. Ytrium spectra obtained by Liquid Scintillation Counting

Fig. 3 presents the spectra obtained by Liquid Scintillation Counting for Sr1 and Sr2 samples. It can be observed that the samples do not contained only Strontium 90 due to the accumulation of Yttrium 90 as a result of Strontium disintegration. The Deconvolution Method was used in order to determine Strontium 90 activity in these samples and eliminate the Yttrium interference. It is an analytical method which allows the measurement of low energy beta emitters in the presence of high energy beta impurities, by spectrum deconvolution [5]. Equations (1) and (2) define the Deconvolution Method mathematical model – Verrezen and Hurtgen (1996).

$$NOI(net,low) = SMP(net,low) - \sum_{k=1}^{n} K_k^{'}(Q,high) \cdot REF_k(net,low) \tag{1}$$

$$K_k^{'}(Q,high) = \frac{SMP(net,high)}{REF_k(net,high)} \tag{2}$$

Where:

– SMP(net,i) – net contribution of the sample (difference between SMP(gross,i) and background BGD(i)) in a given ROI(i).

– NOI(net,*i*) and REF*k*(net,*i*) – net contributions of the nuclide of interest (*i*) and impurity (*k*) (from the reference spectrum in the same region of interest (*i*).
– *low* and *high* indexes represents a ROI in the low and high, respectively, energy range NOI(net,high) = 0 thus, SMP(net,high) = IMP(net,high).

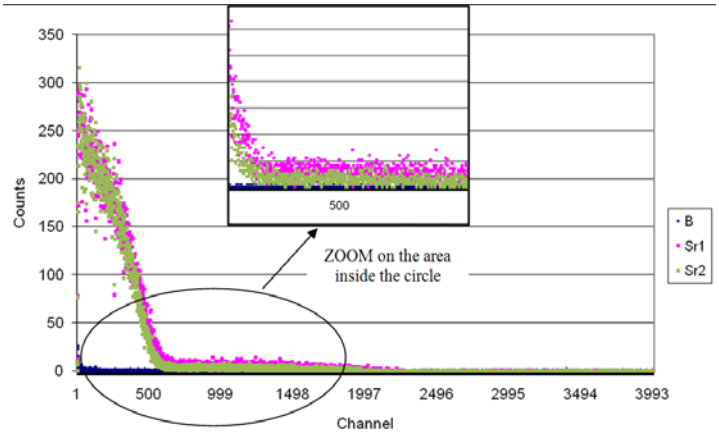

Fig. 3. Spectra obtained for Sr1 and Sr2 Samples by LSC
(B is the spectra for the blank sample)

In Fig. 4 it can be observed the spectra obtained after eliminating the Yttrium interference. Table 2 presents the activity, in Bq, of Strontium 90 in Sr1 and Sr2 samples. To validate the results obtained by using the Deconvolution Method, a correction for the Yttrium accumulation in the Sr1 and Sr2 samples was made by using the decay law. In this case the activity of Strontium 90 in Sr2 sample was 136 Bq, while by using the deconvolution method the resulted activity was 133 Bq, which demonstrates the accuracy of the deconvolution method.

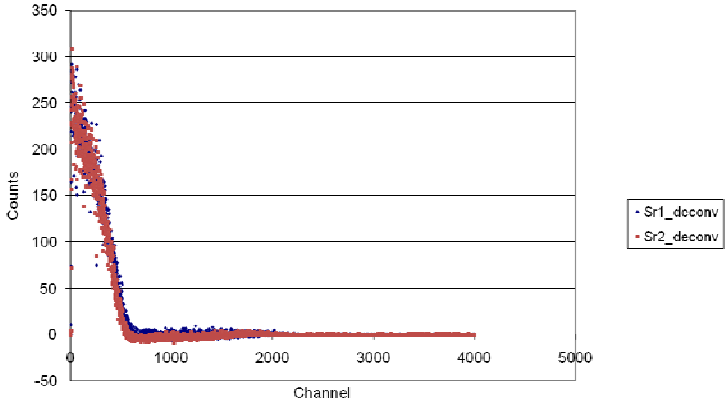

Fig. 4. Spectra of Sr1 and Sr2 Samples after deconvolution

Table 2

Activity of Strontium 90 in Sr1 and Sr2 samples (after applying the deconvolution method)

Sampe No.	Initial solution	Sr1	Sr2
Activity [Bq]	129	136	133

If we consider the uncertainty of the initial solution, uncertainty of the mass when we prepared the solutions and the uncertainty of our final results, we can say that the recovery of Strontium 90 was very good, close to 100%.

4. Conclusions

In Romania, the methodology for a proper radiological characterization of the spent resins from a CANDU 6 NPP has not been established yet and it represents a necessity in order to prepare the radioactive waste packages for final disposal.

The method presented in this paper represents a first step in establishing a methodology for the determination of Strontium 90 in radioactive waste packages, such as spent ion exchange resins, where other interferring radionuclides may be found.

The results obtained for the samples prepared for this paper show that we can continue our work and the next step is to validate the metod on actual spent ion exchange resins samples and also prform the dissolution of these samples.

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