RADIOACTIVE DECONTAMINATION TECHNIQUE USED IN DECOMMISSIONING OF NUCLEAR FACILITIES

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The decontamination of equipment and nuclear installations is a concern from many points of view. The method adopted can minimize the radioactive waste, preventing the environmental pollution and optimizing the environmental protection. For this purpose a research study was developed to evaluate the efficiency of two products, solutions for chemical and radioactive clean-up recently introduced to the market. The ability of the agent to remove the contamination was tested for five types of materials. The objective of the study was to evaluate the deftness of the solutions to remove the contamination with ^{137}Cs and ^{60}Co. The results were quantitative by evaluated using the percent of contamination removal by gamma-ray spectrometry.

Key words: decommissioning, decontamination, gamma-ray spectrometry.

1. INTRODUCTION

In the national and international technical and scientific community, considerable efforts are made for the decommissioning of nuclear facilities to be made in terms of nuclear safety principles that requires organizational and technical measures taken for ensuring the environmental, population and property protection and personnel health, taking into account the costs involved. Numerous challenges are present in the process of radioactive decontamination [1] and decommissioning of the nuclear facilities. In most cases, the decontamination must be non-destructive, the cost must be reasonable, to not inhibit the next activities and the current activity must be safe for all participants and workers. Based on regulatory agencies, many tasks are involved, each of which requires attention to health and safety issues for workers and public, monitoring and management of schedules and costs. The removal of the radioactive contamination which is present on surfaces or spread throughout a work area can be realized through two methods: mechanical or chemical. The chemical methods are usually used for the components and tools decontamination that are immersed in a tank with a chemical solvent to dissolve the contaminant, or by using the polishing techniques to remove

the surface layer. C.H. Jung [2] has reviewed the chemical decontamination processes operating through immersion of components in aggressive chemical solutions. In these applications the chemical decontamination techniques produce large amounts of liquid radioactive waste [3, 4]. Therefore, to avoid the disadvantages of chemical decontamination techniques, but keeping their high effectiveness, it is necessary to develop processes using chemical gels instead of chemical solutions [5]. The chemical gels are used as carriers of chemical decontamination agents. This method is efficient in situations where a longer contact is required, together with the need to minimize the radioactive waste. The chemical decontamination process with gel consists of applying by spraying or brushing large surfaces of components (floors, walls, equipment, etc.) that must be decontaminated.

The aim of this research study is to evaluate the applicability and the functionality of two types of decontamination solutions, DeconGel^{TM}1101 and DeconGel^{TM}1102, manufactured by Cellular Bioengineering, Inc. [6]. According to the product data sheet, both solutions are recommended for the decontamination of radioisotopes as well as particulates, heavy metals, water-soluble and insoluble organic compounds (including tritiated compounds). The gels can be applied to horizontal, vertical and inverted surfaces and can be applied to most surfaces. When dry, the product locks the contaminants into a polymer matrix. The film containing the encapsulated contamination can then be peeled and disposed of according to regulations.

2. EXPERIMENTAL INPUT

Five type of materials were considered in the present study: stainless steel (S), painted tile (P), linoleum (L), unpainted concrete (U) and ceramic (C). These types of material were chosen because they are commonly found in the VVR-S nuclear research reactor building, from Magurele. Two individual sample of 19×12 cm^2 were dedicated for each type of material, each radionuclide and each gel type. The radioisotopes chosen for testing the ability of gels to remove the contamination are the main radionuclide from VVR-S nuclear research reactor: ^{137}Cs (solution with chemical composition CsCl in HCl 0.1 N) and ^{60}Co (solution with chemical composition $CoCl_2$ in HCl 0.1 N). The activity was evaluated using gamma-ray spectroscopy scintillation. The gamma-ray spectrometry system used in the study consisted of a ScintiPack Photomultiplier Base with Preamplifier and High Voltage Supply type 296 and a Digital Portable Multichannel Analyzer type DigiDART. The detector is a 3"×3" NaI(Tl) scintillator, with an energy resolution of 70.62 keV at 1332 keV (^{60}Co), working at +1000 V high voltage.

3. METHOD

The first task was to clean the surface of the sample. After drying, an area of 7.065 cm^2 (a circle with 3 cm diameter) was bordered in the center of each sample. The level of the background radiation was measured before the experiment to start. The radioactive solutions were spread on the 7.065 cm^2 surface of the sample, using a single channel pipette; then, were placed in a ventilated hood for 24 hours for drying. Additionally, lubricant was applied on surface samples used in the decontamination experiments with DeconGel^{TM}1102. In the next step, the measurements were performed on materials contaminated with ^{137}Cs and ^{60}Co radioactive solutions with the gamma-ray scintillator. The first decontamination was performed applying the decontamination gels with a device that ensure the uniformity of the gel thickness of approximately 0.3 mm over an area of 6.5×6.5 cm^2-larger than the surface with radioactive solution. After 24 hours, the gel was completely dried and could be peeled as a continuous sheet being kept between two sheets of paper. Activity measurements were performed on the sample surface decontaminated and on the peeled gel.

For the evaluation of the activity measurement, the detection efficiency was evaluated with NaI(Tl) detector [7, 8]. The procedure was complicated due to the presence of important coincidence summing effects in the case of ^{60}Co radionuclide. Dedicated software called GESPECOR [9] was used in order to evaluate the coincidence summing corrections presented.

A second decontamination was performed using the same type of gels involved in the study, for each sample.

4. RESULTS

The background was measured and considered for each type of material without radioactivity solutions. Firstly, the level of initial activity measured on the contaminated sample with ^{60}Co (the first and second sample from each type of material) and ^{137}Cs (the 3rd and 4th sample from each type of material) was evaluated for both DeconGel^{TM}1101 and DeconGel^{TM}1102 experiments. In Fig. 1 are represented the values of the initial activity for materials involved in the experiment with DeconGel^{TM}1101 and DeconGel^{TM}1102.

Then, the levels of the activity incorporated into DeconGel^{TM}1101 and DeconGel^{TM}1102 and the activity remaining on sample material, after the first and second decontamination were evaluated through gamma-ray spectrometry.

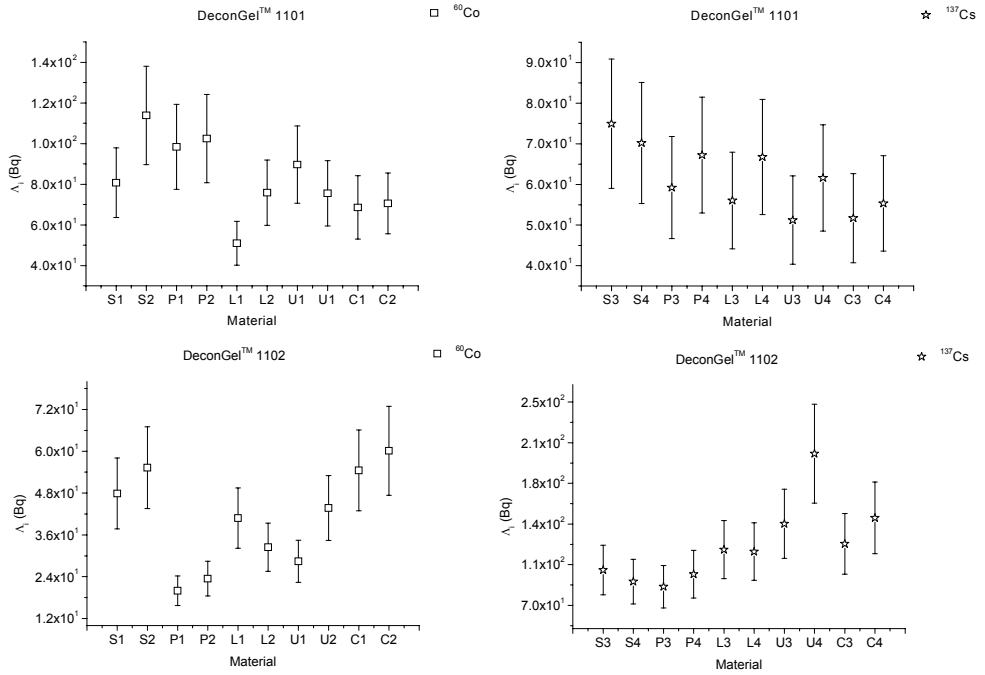

Fig. 1 – The initial activity for experiment with DeconGel™1101 and DeconGel™1102.

5. ANALYSIS AND DISCUSSIONS

In order to quantify the ability to remove the contamination using both DeconGel™1101 and DeconGel™1102, the decontamination factor was evaluated for the materials involved in the present study, using two methods:

$$DF1(\%) = \frac{\Lambda_i - \Lambda_{m.I}}{\Lambda_i} * 100 \quad (1)$$

were: Λ_i is the initial activity; $\Lambda_{m.I}$ is the remaining activity on the sample, after the first decontamination evaluated through gamma-ray spectrometry [7]

$$DF2(\%) = \frac{\Lambda_{g.I}}{\Lambda_i} * 100 \quad (2)$$

were: Λ_i is the initial activity; $\Lambda_{g.I}$ is the activity incorporated in gel, after the first decontamination evaluated through gamma-ray spectrometry.

To assess the decontamination factors, the results obtained by gamma-ray spectrometry measurements after the first decontamination were used, because the activities results obtained after the second decontamination were below the minimum detectable activity. This indicates a high efficacy of decontamination using chemical gels. To represent graphically the decontamination factor, an average value of the *DFm* was express averaging the values of the decontamination factors obtained for the two samples (after the first decontamination with DeconGel^{TM}1101 and DeconGel^{TM}1102) for each radionuclide and material. In Fig. 2 are presented the average decontamination factor values for all materials involved in the study, for each radionuclide measured (after the first decontamination) through gamma-ray spectrometry measurement with NaI(Tl) scintillator.

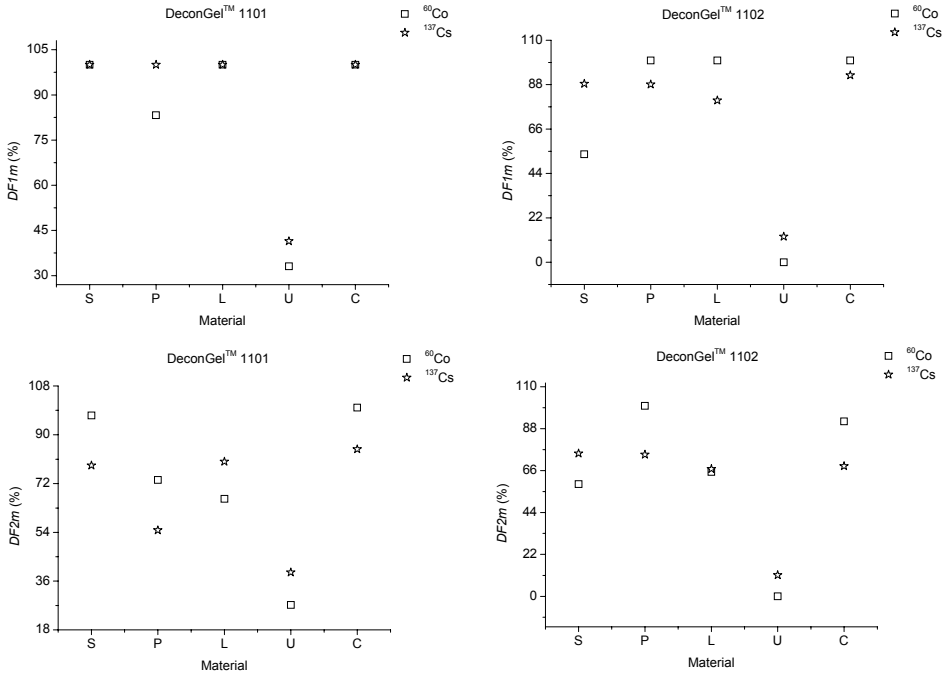

Fig. 2 – The average decontamination factor for ^{60}Co and ^{137}Cs.

The two methods used in the study to calculate the decontamination factor shows a different perspective of the process to remove the contamination. The percentage of contamination removed from the material evaluated with the first method (eq. (1)) shows the efficacy in decontamination process with DeconGel^{TM} 1101 and DeconGel^{TM} 1102. In the case of DeconGel^{TM} 1101 the percentage of

contamination removed from the material varies in descending order from stainless steel (S), linoleum (L), ceramic (C), painted tile (P) and unpainted concrete (U) for ^{60}Co; and from stainless steel (S), painted tile (P), linoleum (L), ceramic (C) and unpainted concrete (U) for ^{137}Cs. The efficacy in decontamination process using DeconGel^{TM} 1102 varies regressively as follows: painted tile (P), linoleum (L), ceramic (C), stainless steel (S) and unpainted concrete (U) for ^{60}Co; and ceramic (C), stainless steel (S), painted tile (P), linoleum (L) and unpainted concrete (U) for ^{137}Cs. The percentage of the activity incorporated into the gel evaluated with the second method (eq. (2)) shows how effective are DeconGel^{TM} 1101 and DeconGel^{TM} 1102 to isolate and encapsulate the contamination present on the material sample. In the case of DeconGel^{TM} 1101 this percentage was highest for ceramic (C) then for stainless steel (S), painted tile (P), linoleum (L) and unpainted concrete (U) for the case of ^{60}Co; and for ceramic (C), linoleum (L), stainless steel (S), painted tile (P) and unpainted concrete (U) for the case of ^{137}Cs. The effective of DeconGel^{TM} 1102 to isolate and encapsulate the contamination present on the material sample was variable in regressive sequence from painted tile (P), ceramic (C), linoleum (L), stainless steel (S) and unpainted concrete (U) for ^{60}Co; and from stainless steel (S), painted tile (P), ceramic (C), linoleum (L) and unpainted concrete (U) for ^{137}Cs. Was observed that the efficiency of decontamination with chemical gels depend on material type and radionuclide. For all the unpainted concrete (U) samples was recorded the lowest level of decontamination. This behavior is due to the fact that the concrete is a porous material and as a consequence the radioactive solutions were infiltrated in the sample volume. A chemical reaction can be caused by deeper infiltration of the radioactive solution in the volume material. Those actions may result in decrease of the contamination removal degree [10, 11].

Due the additional errors composed of systematic and random errors or due to loss of particle material during the removal contamination process, the radioactivity that was either “lost” or “found” was observed. Because the decontamination factor evaluated through the first method allows a more realistic comparison of data than the second method that represent an alternative to demonstrate the percent incorporated in the gel, first method was chosen the evaluate the percentage of this “lost” or “found” radioactivity after all measurements were completed (Fig. 3).

From the representation was obvious that the highest percent of the “lost”/“found” radioactivity was in the case of unpainted concrete (U). In addition, in the case of these samples, was observed that after gels application and drying, this was difficultly exfoliated although a utility knife for peeling off was used and a tweezers for removal processes. In this case, adding an extra layer of gel resulted in an easy separation gel film.

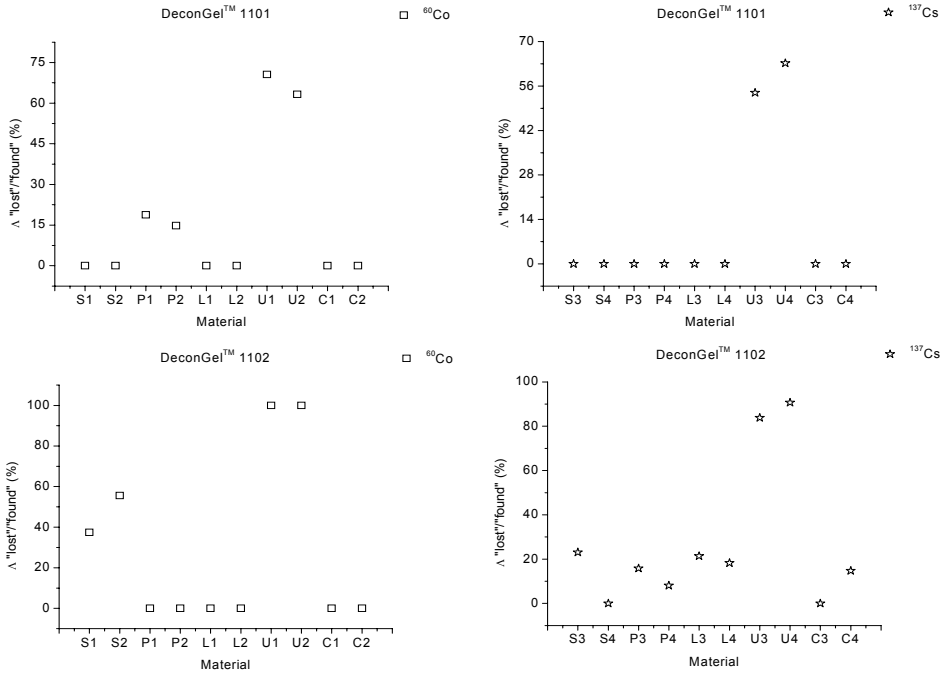

Fig. 3 – The percentage of radioactivity "lost" or "found".

6. SUMMARY AND CONCLUSIONS

The DeconGel™1101 and DeconGel™1102 involved in this study have shown high efficiency in the contamination removal experiments for the stainless steel (S), painted tile (P), linoleum (L), unpainted concrete (U) and ceramic (C) samples. These types of gels can be applied to horizontal, vertical and inverted surfaces and can be applied to most surfaces and operates by dissolving the radioactive deposition, together with a thin layer of gel support, so the radioactivity trapped in the surface can be removed. After drying, this solution forms a strong film that can be peeled from the surface. Only small amount of gel are needed, and the polymer will show the thixotropy properties, *i.e.* they are in liquid form when are applied, and solid when stationary, allowing strong adhesion to surfaces. The practical experiments made with DeconGel™1101 and DeconGel™1102 respectively, showed that the decontamination process may cause acceptable results and reasonable cost. In experiments was not emphasized any detectable exposure of operating personnel associated with decontamination operations, reducing the risk level of operating personnel involved in the operations. DeconGel™1101 and DeconGel™1102 present the following advantages: zero preparation, easy application, simply decontamination, minimal odor (not require

the use of protective masks). This type of gel can be considered an effective means to reduce or eliminate the surface contamination from systems of buildings or equipment. This technique decontamination can improve worker safety and reduce requirements for personal protective equipment, resulting in reduced costs and possible acceleration of nuclear facility decommissioning process.

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