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EFFICIENCY ASSESSMENT OF SANITARY-TECHNICAL BARRIERS IN NUCLEAR FACILITIES BY MEANS OF ADHESIVE DECONTAMINATING FOILS

Tzvetan Marinov[*,], Hristo Protohristov[**], Nikolay Goutev[**], Dobromir Dimitrov[**], Alexandar Nikolov[**], Nasko Mihov[**], Anguel Demerdjiev[**], Elena Geleva[**], Dimitar Tonev[**,#]**

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Abstract

The efficiency of sanitary-technical barriers in Kozloduy NPP, Bulgaria, is investigated for the first time with adhesive decontaminating foils. The duration of use and retention coefficient are assessed using gamma-ray spectroscopy. The retained activity increases linearly up to a saturation point at 250 controlled persons. About 6% of the radioactive mechanical fraction is fixed on the adhesive. For three successive sanitary-technical barriers in the controlled zone, the activity is reduced by one to two orders of magnitude on each barrier towards the sanitary complex exit. The total activity released during annual maintenance is estimated to be 10^5 Bq. The radioactive contamination in the controlled zone is studied and correlations are established between two groups of isotopes, generated by the release of fission products and by neutron activation. Our investigation demonstrates that the sanitary technical barriers prevent efficiently the release of radioactivity from the controlled zone to the environment.

Key words: radioactivity, gamma-ray spectroscopy, adhesive decontamination foil, whole-body monitor, nuclear power plant

[#]Corresponding author.

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Introduction. Nuclear power plants (NPPs) and other nuclear facilities are the main sources of artificial radioactivity in the controlled zone (CZ), in the observation zone around the NPP, and in the environment, where the radiation dose to the population can be eventually influenced. Successive sanitary technical barriers (STBs) are established in the NPP to reduce the radioactive emissions below the prescribed limits. Gaseous fission products generated during operation of Kozloduy NPP, Bulgaria, are removed by radiochromatographic absorption in active carbon filters. Their efficiency has been investigated previously [1]. Personnel and equipment are controlled at the STBs by means of whole-body radiation monitors (WBRMs) [2] and in certain cases the controlled objects have to be decontaminated until the radioactive contamination is reduced below the threshold level of the respective barrier. Different types of radioactivity (surface alpha- and beta-, as well as gamma-radiation) are measured at all checkpoints of the consecutive barriers. The results of the measurements, together with the personal identification numbers, are registered and stored in protocols. From these data, the efficiency of each STB can be roughly estimated, but a systematical investigation of the efficiency of STBs requires more advanced methods with higher sensitivity to be developed for precise assessment of a single STB and for assessment of the whole barrier system in the NPP. At present, the study of surface contamination in different facilities of the CZ is limited to collection and measurement of samples smeared with tampons from surfaces and successive measurement of the alpha- and beta-, as well as gamma-radiation activity with radiometers and spectrometers [3]. Disadvantages of this method are the time-consuming procedure, the small control area (100 cm^2) and subjective factors. Here, for the first time, a method for precise efficiency assessment of STBs is applied, based on high-resolution gamma-ray spectroscopy measurements of adhesive decontaminating foils. The method allows detection of specific contamination sources. The paths of diverse radionuclides can be identified and correlations between them can be extracted; the history of contamination events can be recovered and by their analysis the efficiency of all STBs could be improved. Adhesive foils have been successfully applied on sanitary barriers for reduction of biological contamination in clean rooms and biolaboratories, and their efficiency has been investigated by measuring the concentration of pathogens removed from the shoe soles into the foil [4].

Experimental method. Adhesive foils are generally used to reduce the spread of dust and small particles carried on shoe soles in clean rooms, laboratories, etc. In nuclear facilities adhesive foils are often used for partial decontamination of small radioactive mechanical fraction. The project of the National Cyclotron Centre in Sofia, Bulgaria, foresees the use of adhesive mats at the entrances of the clean rooms [5,6]. The experimental approach reported in the present paper is applicable not only for NPPs, but also for accelerator centres with radiopharmaceutical production. Commercially available decontaminating mats are used for this purpose in Kozloduy NPP.

The decontaminating mat consists of up to 60 foils, coated with a sensitive adhesive, with dimensions 117 × 91 cm. The adhesive collects dust and small particles from the shoes and from the wheels of service carts. For our experiments, the mat is placed directly in front of and on the exit of the WBRM. When the top foil is worn out, it is peeled off to expose the next clean foil ready for use, and so on. The used foils are labelled with a number, time, duration of use, number of controlled persons passed through the WBRM, etc., and brought for laboratory investigation.

Gamma-spectroscopy system and measurements. The gamma-ray measurements were performed on a high-resolution gamma-spectrometer equipped with Canberra HPGe detector type GC5019 with the following characteristics: relative efficiency 50%, energy resolution 0.8 keV at 122 keV (^{57}Co) and 1.9 at 1332.5 keV (^{60}Co); photopeak to Compton ratio 64:1. For data acquisition a standard multichannel analyzer with dead time and pileup corrections is used.

The used adhesive foils were placed in a 2-liter Marinelli beaker. During the measurements the detector with the Marinelli beaker was enclosed in a standard passive low-background shielding. The typical measurement time for one foil was 2 h.

The activity of each detected radionuclide was determined by means of its most intensive gamma-ray transition: ^{51}Cr (320.1 keV), ^{54}Mn (834.8 keV), ^{58}Co (810.8 keV), ^{59}Fe (1099.3 keV), ^{60}Co (1332.5 keV), ^{95}Nb (765.8 keV), ^{95}Zr (756.7 keV), ^{110m}Ag (884.7 keV), ^{113}Sn (391.7 keV), ^{124}Sb (722.8 keV), ^{125}Sb (427.9 keV), ^{134}Cs (604.7 keV), ^{137}Cs (661.6 keV), ^{144}Ce (133.5 keV). The net photopeak area was calculated and corrected for the detector efficiency by means of LabSOCS (Laboratory SOurceless Calibration Software) software package [7].

Measurements with WBRMs. The WBRMs are used to control the personnel when crossing the STBs that divide different facilities in the controlled zone. The WBRM measures the beta-/gamma- contamination of human body, hands, clothing and shoes by means of 27 detectors. The data for radioactivity on shoe soles, essential for the investigation, is collected from the two bottom detectors for the left and the right shoe, respectively. The WBRM detectors are calibrated with a ^{60}Co radiation source, and the relative efficiency for ^{60}Co is therefore equal to 1. To compare the results from the gamma-ray adhesive foil measurements with the WBRM results, the activities of certain detected radionuclides have to be equalized through their ^{60}Co equivalents.

Results and discussion. ***Decontamination characteristics of the adhesive foil.*** The experiments were performed during a planned annual maintenance (PAM). On the exit of the CZ an adhesive mat consisting of up to 60 layers of adhesive decontaminating foils was set up in front of the WBRM. When crossing the STB, part of the radioactive mechanical fraction (dust and small particles) on shoe soles sticks to the adhesive foil and remains there. The foil dimensions (117×91 cm) allow successive stepping with both, left and right feet. After a cer-

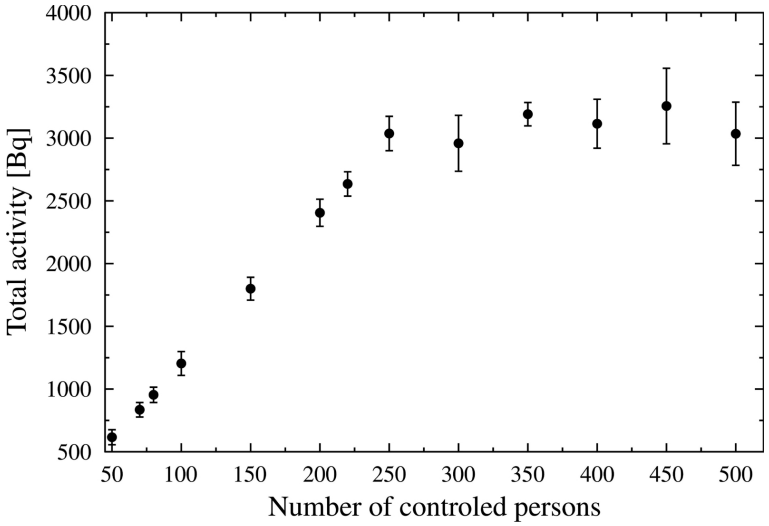

Fig. 1. Graphic plot of the total retained radioactivity on adhesive foils as a function of the number of controlled persons

tain number of persons have passed through the STB, the adhesive foil is peeled off from the mat and its activity and isotopic composition is measured by means of high-resolution gamma-spectrometer.

The overall performance of the adhesive foil was assessed, based on the activity measurements of foils with different utilization rate (number of persons passed through). Figure 1 shows the retained activity as a function of the number of controlled persons.

The activity increases linearly reaching saturation at about 250 controlled persons. Above this number, due to wear and saturation of the adhesive foil, the activity remains nearly constant at about 3 kBq. From the retained activity we can calculate roughly the retention efficiency of the used foil, defined as: $\mathrm{Ks} = \mathrm{A_p}/\mathrm{A_0}$, where, $\mathrm{A_p}$ is the activity retained on the foil and $\mathrm{A_0}$ is the total activity carried on shoe soles. From the linear part of the plot we conclude, that the retention efficiency remains nearly constant in the interval 1–250 controlled persons.

To calculate precisely the retention coefficient of the adhesive foil we combined the results from the gamma-ray spectrometry measurements with the corresponding data from the WBRMs. The analysis of the data collected during the annual maintenance at the exit of the hermozone (hermetical confinement of the reactor vessel and its auxiliaries), considered as the main source of contamination in the CZ, shows that 6.2% of the radioactive contamination from dust and small particles is retained by the foil. The average collected contamination per person is about 40–45 Bq.

The remaining radioactivity is contained and removed by changing clothes, washing, and bathing. Thus, the foils serve primarily as a sampling and monitoring

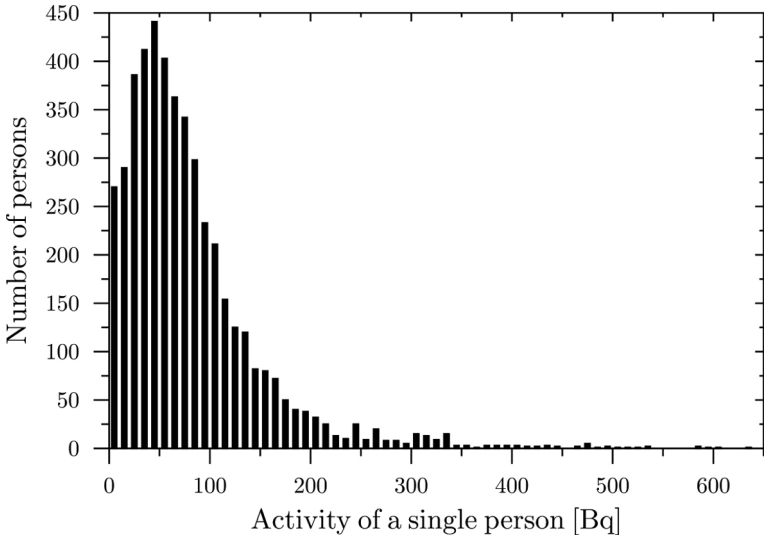

Fig. 2. Histogram of the activity level for a single person passing the last STB at the exit of the CZ

tool with high sensitivity (due to the longer accumulation and better statistics) for evaluation of the STB efficiency against contamination transfer by the shoes of the personnel. The latter is well-known to be the major source of contamination in clean rooms and aseptic production areas, where the adhesive foils are frequently used as a contamination barrier.

The control data for a single person from the WBRM allow statistical study of the distribution of radioactivity carried out from personnel on shoe soles through the STB. Figure 2 shows the corresponding histogram for the last STB at the exit of the CZ.

Here, a nearly normal distribution was observed with a maximum at about 45 Bq. The probability of a person carrying out more than 350 Bq is less than 1%, which demonstrates the high impermeability of the STBs against radioactive contamination.

The pilot tests on different barriers in the CZ during the PAM in 2016 show that the activity is reduced by 1–2 orders of magnitude on each successive barrier starting from the hermozone to the sanitary complex exit. Correlations between radioisotopes have been established, dominated by two groups of nuclides originating from fission products and from neutron activation of construction materials.

Efficiency assessment of the STBs. The efficiency assessment of the STBs has been performed on several STBs in the CZ, during PAMs of 5th and 6th Units of the Kozloduy NPP. The points are specially selected to control the major sources of contamination and the basic personnel flows.

Adhesive decontaminating foils are positioned in front of the WBRM at the exits of the hermozone, CZ, the bath and the sanitary complex. Figure 3 shows

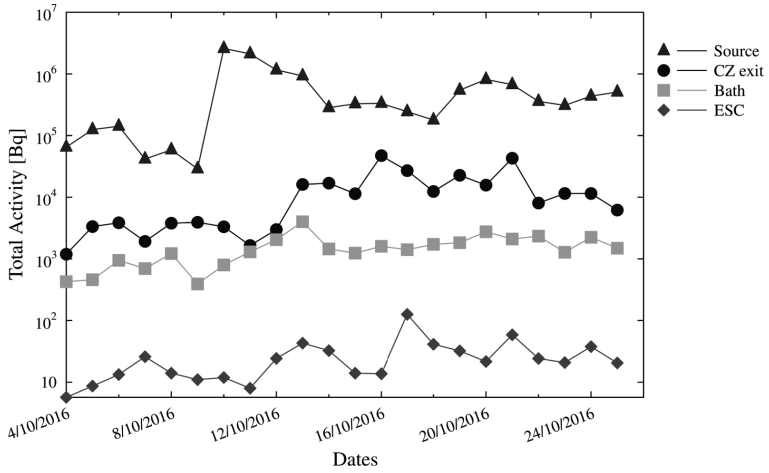

Fig. 3. Total activity levels, measured by means of adhesive foils, on different STBs during the PAM

the total activity collected in these points during the period of the PAM. As expected, the highest activity in the interval of 10^5–10^6 Bq is measured at the exit of the hermozone (source), where the basic activity with contaminated equipment during the PAM is performed. At the exit from the CZ the activity decreases by two orders of magnitude. At the next point (exit from the bath) the activity decreases again by roughly 10 times and at the exit from the sanitary complex (ESC) – by further 100 times.

Based on the measurements at the exit of the sanitary complex and the number of controlled persons, the total activity released from the controlled zone during the PAM-2016 is estimated to be $A_{sum} = 1 \times 10^5$ Bq. This result demonstrates that the STBs are effectively blocking radioactive contamination of the environment.

Considering the efficiency assessment of the STBs, some advantages of the new method are mentioned compared with the conventional smear sampling method. The activity concentration of a smear sample is about 40 Bq under the following conditions: surface contamination 4 Bq/cm^2, average take-off rate 10% and smear sampling area 100 cm^2. Here, a relatively small fraction of the activity is glued to the floor and therefore the method of evaluation by smear sampling is limited in reliability and accuracy.

In comparison, the activity concentration on the adhesive foil is about 2000 Bq for 250 controlled persons with an average contamination of 0.5 Bq/cm^2 per shoe and a 6% take-off rate. The new method reflects the mechanism of spreading pollution through work shoes by transporting it from the radioactivity source to the exit of the controlled area. It makes possible to quantify the average contamination of work shoes, which is an important parameter for establishing a

histogram of the distribution of individual contamination (Fig. 2) and cannot be extracted by means of smear sampling.

Correlation analysis. Correlation analysis is performed to determine the interconnections between activity levels of different radionuclides, detected at the STBs by means of adhesive decontaminating foils. This analysis shows how much a change in the activity of a certain radionuclide is connected with a change in the activity of other radionuclides. EXCEL Data Analysis software was used for the calculations. The correlation strength is expressed by the correlation coefficient which varies from 0 to 1. For coefficient values in the interval 0.0–0.3 there is no correlation; from 0.3 to 0.7 there is a weak correlation; in the interval 0.7–1.0 a strong correlation is observed. Table 1 presents the results of the correlation analysis.

T a b l e 1

Results from the correlation analysis of activity levels measured at the different STBs by means of adhesive decontamination foils

Isotope	^{51}Cr	^{54}Mn	^{58}Co	^{59}Fe	^{60}Co	^{95}Nb	^{95}Zr	^{124}Sb	^{125}Sb	^{137}Cs	Sum
^{51}Cr	1.00										
^{54}Mn	0.83	1.00									
^{58}Co	0.94	0.87	1.00								
^{59}Fe	0.69	0.63	0.69	1.00							
^{60}Co	0.48	0.76	0.50	0.32	1.00						
^{95}Nb	0.14	0.13	0.12	0.06	0.09	1.00					
^{95}Zr	0.16	0.14	0.12	0.04	0.11	1.00	1.00				
^{124}Sb	0.57	0.51	0.58	0.61	0.39	0.25	0.23	1.00			
^{125}Sb	0.41	0.36	0.33	0.33	0.45	0.46	0.45	0.73	1.00		
^{134}Cs	0.23	0.16	0.18	0.08	0.22	−0.04	−0.04	0.31	0.45	1.00	
Sum	0.97	0.91	0.94	0.68	0.64	0.28	0.30	0.61	0.50	0.23	1.00

A strong correlation is observed between the radionuclides ^{51}Cr, ^{54}Mn, ^{58}Co and ^{59}Fe, which could be explained by their common origin from neutron activation of the construction materials of the nuclear reactor and its auxiliary systems.

The correlation between ^{60}Co and the group ^{51}Cr, ^{54}Mn, ^{58}Co, ^{59}Fe is of medium strength, which can be explained by independent contamination sources of ^{60}Co. These sources could be contamination from previous maintenance in the "dirty workshops" (workshops, where equipment from the hermozone is repaired). ^{51}Cr, ^{54}Mn, ^{58}Co and ^{59}Fe have shorter half-lives than ^{60}Co ($T_{1/2} = 5.26$ years), therefore their activity decreases considerably with time, but ^{60}Co can be spread within the respective zones for a longer period and will dominate the contamination.

No correlation was observed between fission products (e.g. ^{137}Cs) and other radionuclides, which shows that the corresponding contamination originates from previous PAMs, when partial dehermetization (clad failures) of fuel elements occurred in the primary circuit.

Conclusions. Efficiency assessment of STBs in Kozloduy NPP using adhesive decontamination foils has been carried out. The overall performance and the deposition coefficient of the adhesive foil for mechanical radioactive contaminants are determined by means of high-resolution gamma-ray spectrometry. The deposited activity increases linearly reaching saturation at about 250 controlled persons and a total value of about 3 kBq, whereas about 6% of the radioactive contamination remains on the foil. The average contamination of a person is estimated to be 40–45 Bq. The test on three STBs in the CZ during a PAM shows that the activity is reduced by 1 to 2 orders of magnitude on each successive barrier starting from the radioactive source (hermozone) to the sanitary complex exit. The total activity released from the CZ during a PAM is estimated to be 10^5 Bq. This result demonstrates that the STBs are effectively blocking the release of radioactivity to the environment.

Correlations between radionuclides have been established, dominated by two groups of isotopes originating from fission products and from neutron activation of construction materials. The obtained original results are very important for the safe and efficient decommissioning of nuclear facilities.

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*Kozloduy NPP PLC
3321 Kozloduy, Bulgaria

**Institute for Nuclear Research and Nuclear Energy
Bulgarian Academy of Sciences
72 Tsarigradsko Shosse Blvd
1784 Sofia, Bulgaria
e-mails: tsvmarinov@abv.bg
proto@inrne.bas.bg
nikolay.v.goutev@hotmail.com
dobri@inrne.bas.bg
artynik@abv.bg
nmihov2002@yahoo.co.uk
a.demerdziev@gmail.com
lenag@inrne.bas.bg
dimitar.tonev@inrne.bas.bg