

COMPARISON OF DECONTAMINATION STANDARDS

LE CONG HAO,¹ MAI DINH THUY,² DO TRUNG HIEU,³ AND ZOLTÁN SAS^{4*}

¹ Nuclear Techniques Laboratory, University of Science, VNU-HCMC, Ho Chi Minh City, VIETNAM

² School of Nuclear Engineering and Environmental Physics, Hanoi University of Science and Technology, Ho Chi Minh City, VIETNAM

³ Faculty of Chemistry, University of Science, VNU, Ho Chi Minh City, VIETNAM

⁴ Institute of Radiochemistry and Radioecology, University of Pannonia, Veszprém, H-8201, HUNGARY

The quality of materials used in nuclear-related facilities is critical, especially the ease of decontamination of different paints and coatings. Standards describe different testing methods for classification. Nevertheless, compliance with these standards cannot be carried out negligibly from a safety point of view. In this study, a withdrawn Hungarian (MSZ-05 22.7662-83), an international ISO (ISO 8690:1988), and Russian (GOST 25146-82) decontamination standard were compared. Four different paints were tested as part of this survey. The ease of decontamination varied mainly from poor to fair levels in the case of the Hungarian standard, while the ISO standard exhibited very good level. In the case of the Russian standard, only a theoretical comparison was carried out. Based on the results, it was found that a special epoxy-based coating can be recommended for isotope laboratories due to being the best material from an ease of decontamination point of view. From comparison of the standards considered here, it was found that the application of ISO standard is significantly faster and simpler than the withdrawn Hungarian standard. However, in the case of the Hungarian standard the data described the ease of decontamination in more details. The use of water or some other cleaning agents can be effective to remove ¹³⁷Cs and ⁶⁰Co contamination right after early identification. Isotope ¹³⁷Cs and ⁶⁰Co contamination of a surface can be cleaned quickly and effectively using distilled water for the ¹³⁷Cs isotope removal from the surfaces being several times easier than that of ⁶⁰Co.

Keywords: surface contamination, decontamination value, ISO standard 8690:1988, Hungarian standard MSZ-05 22.7662-83, Russian standard GOST 25146-82, ⁶⁰Co isotope, ¹³⁷Cs isotope

1. Introduction

Contamination of surfaces with radionuclides can lead to human exposure depending on the type, extent of contamination, and activity of the contaminating isotope. In order to reduce the risk, quick and effective decontamination of the involved area is required. The contamination of different surfaces are common in workplaces that deal with radionuclides e.g. isotope laboratories, nuclear industry related activities, etc.[1,2]. Contamination can occur in various ways, but chemical and physical adsorption processes are the most important ones. In the case of chemical adsorption, ions exit from the hydration shell and bind directly to the surface, while in the case of physical adsorption the ions binds to the surface together with the hydration shell. Furthermore, the contaminating isotopes can infiltrate into the pores of the surface material *via* diffusion. To avoid the internal contamination of porous materials special paints and coatings should be applied which inhibits the diffusion of contaminating isotopes into

pores [3]. The decontamination capacity of surfaces greatly depends on the form of contamination media and the chemical and physical parameters of surfaces. The main influencing parameters that can affect the ease of decontamination are i) surface porosity, ii) surface roughness, iii) surface wettability, iv) chemical reactions between the radionuclide and the surface, and v) adsorption processes on the outer part of the electric double layer of the solid/liquid interface.

The efficiency of decontamination expressed by the decontamination factor (*DF*) [3] can be calculated according to the following equation:

$$DF = \frac{\text{Activity of surface after contamination}}{\text{Activity of surface after decontamination}} \quad (1)$$

However, the decontamination efficiency is greatly parameter dependent as mentioned above. Standardised protocols are necessary to classify certain paints and coatings from a decontamination point of view, which is informative about their utilisation in isotope laboratories and other relevant workplaces as well.

In this study, the comparison of different decontamination standards is presented that includes a Hungarian (withdrawn in 2003) standard MSZ-05 22.7662-83 “*Testing of painted coatings in laboratory.*

*Correspondence: ilozas@almos.uni-pannon.hu

Table 1. Properties of the coatings investigated.

ID	coating type	paint	colour	roughness
DC	base-modified silicone resin	spray	grey (Fig.1A)	slight
CV	alkyd resin	spray painted	dark grey (Fig.1B)	great
KM	enamel paint	brush painted	brown (Fig.1C)	glossy
NR	epoxy resin	brush painted	beige (Fig.1D)	semi-glossy

Determination for ease of decontamination”[3], an international ISO 8690:1988 standard “Decontamination of radioactively contaminated surfaces – Method for testing and assessing the ease of decontamination” [4], and a Russian interstate standard GOST 25146-82 “Radiochemical production and atomic power plant materials. Method for determination of decontamination ratio” [5].

2. Experimental

2.1. Sample Preparation

In order to compare the three selected standards, four different types of coatings were used for providing various conditions during the survey. The relevant properties of the applied coatings are shown in Table 1.

The selected coatings were painted on 4×4 cm aluminium test disks (Fig.1) and stored for 24 hours to dry completely. The selected standards require 5 parallel measurements for each type of coating.

2.2. Comparison of Contamination Processes

In the case of the Russian GOST 25146–82 standard, the investigated surfaces are contaminated by natural or artificial β -radiating nuclides. The activity of the samples was distributed on the surface to avoid self-absorption. The GOST standard describes in great detail the measurement conditions and apparatus, which should be enforced to ensure reliable results. The same standard allows for using any types of contamination solution, which provides numerous ways for measuring the ease of decontamination under a wide variety of conditions, while in the case of the ISO and MSZ standards specific conditions have to be maintained. In the case of the withdrawn Hungarian MSZ-05 22.7662-83 and ISO 8690:1988 standards the recommended contaminating isotopes are ^{137}Cs and ^{60}Co (carrier concentration $10^{-5} \text{ mol dm}^{-3}$ and pH value of 4) which were prepared in a laboratory before testing.

To determine the count rate of the contaminating isotopes a γ -spectrometer was used with a high purity germanium (HPGe) semiconductor detector ORTEC GMX40-76, with 40% efficiency. To obtain counts from ^{137}Cs , the 661.6 keV γ -line was measured, while in the case of ^{60}Co , the 1173.4 and 1332.5 keV lines were

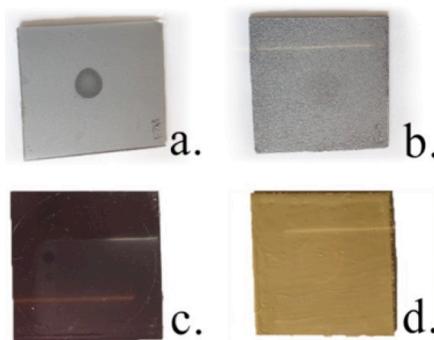


Figure 1. Applied coatings on test disks.

measured. The spectra were recorded by an ORTEC DSPEC LF 8196 MCA instrument. Before contamination the background spectra were recorded, which were extracted from all contaminated and decontaminated spectra.

2.3. Hungarian Standard MSZ-05 22.7662-83

For the Hungarian standard, the decontamination was investigated in two differentiated ways to obtain relevant information related to physical and chemical links adsorption separately. To investigate the physical adsorption, 0.1 cm^3 of contamination solution was dropped onto test samples and dried under an infrared lamp at $40 \text{ }^\circ\text{C}$. After drying, the count rates were measured using a HPGe detector for 1000 s to obtain the specific count rate of the contamination solution. To investigate chemical origin contamination, a special socketed cylinder-shaped contamination block (Fig.2) was used with 0.565 cm^3 of the contamination solution, which provides a 10 cm^2 contact surface between the solution and test specimens. The contamination block was put onto coated test specimens and filled with the contamination solution for 2 hours. After that period, the contamination solution was removed from the contamination block and the test specimens were gently flushed using ultrapure water.

2.4. ISO Standard 8690:1988

For the ISO standard, only chemical adsorption was investigated. The specific count rate of the contamination solution was determined before contamination of the test specimens. A micropipette was used to put 0.1 cm^3 of contamination solution onto glass sheets. The test specimens were inserted between the upper and lower parts of the contamination block for contamination, which was prepared according to the ISO standard (Fig.3).

Before filling the upper part with 1 cm^3 of contamination solution both parts were fastened together tightly to avoid leakage. After filling the holder, disks were covered to avoid evaporation of the contamination solution for 2 hours. After this contamination process, the contamination solution was pumped out of the holder, and the test specimens inserted into the decontamination unit.

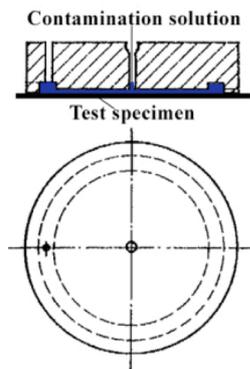


Figure 2. Scheme of the contamination block according to the withdrawn Hungarian MSZ-05 22.7662-8 standard.

2.5. Decontamination Process

In the case of the Hungarian standard, an immersion-based decontamination method was used over three steps. Firstly, upon the completion of count rate measurements, the contaminated surfaces were immersed in ultrapure water for 10 s then pulled out and tilted to allow the residual fluid to trickle down before finally being immersed again in the decontamination solution. The immersion was repeated 15 times (total immersion time: 150 s). Thereafter, the test specimens were dried under an infrared lamp and the count rate originating from the residual contaminating isotopes recorded using a γ -ray spectrometer.

In the second step, a special decontamination solution was prepared according to the method described as a standard in order to get information about the decontamination efficiency of detergents. The decontamination solution was composed of polyethylene glycol nonylphenyl ether (5 g dm^{-3}), citric acid (4 g cm^{-3}), and EDTA (4 g cm^{-3}). The test specimens were added to the decontamination cocktail and decontaminated using the same immersion/pull out technique described in the first step. The count rate was measured again after drying. For the final step 1 M HCl was used to get information about more aggressive decontamination fluids, which can cause structural changes in the case of the investigated coating but can also be beneficial from a decontamination point of view to remove isotopes from pores also. The decontamination and the measurement process were repeated for each specimen after the acidic decontamination step.

In the case of the ISO standard, the test specimens were placed immediately into a special cage-stirrer apparatus (described in the standard) after the contamination process. The apparatus was equipped with a 100 rpm motor. The cage was immersed into a glass beaker filled with ultrapure water and rotated for 150 s. Thereafter, the specimens were dried and measured using γ -ray spectrometry to obtain residual count rates. The details of the MSZ-05 22.7662-83 and ISO 8690:1988 decontamination methods are summarised in Table 2.

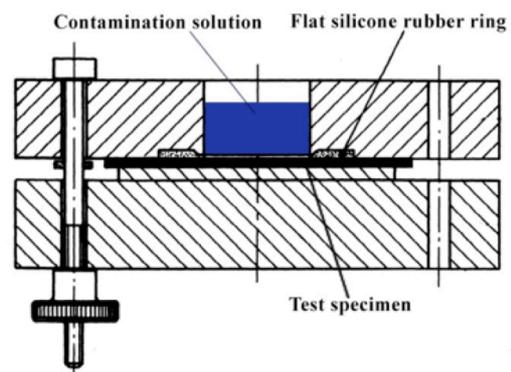


Figure 3. Scheme of the contamination block according to the ISO 8690:1988 standard.

Table 2. Results of decontamination experiments performed in this study.

standards	steps	method	agents
MSZ-05 22.7662-83	3	immersed for 150 s	ultrapure water decontamination solution
MSZ-05 22.7662-83 adsorption		then pulled out	1 M HCl
ISO 8690:1988		1	immersed stirring cage in ultrapure water water for 150 s

2.6. Calculation of DF Values and Classification of Specimens

The decontamination factors were calculated from recorded spectra. The peak areas corresponding to the presence of ^{137}Cs and ^{60}Co isotopes were corrected by background measurements. The specific count rates (count-per-seconds cm^{-3}) were calculated for all samples. The decontamination factors of each step for all samples were calculated using the Eq.(1).

3. Results and Discussion

3.1. Decontamination Factors from the Hungarian Standard MSZ-05 22.7662-83

The obtained decontamination factor of each treated surface on the basis of the Hungarian standard is shown in Fig.4, which compares the physical adsorption between the contaminating isotopes and surfaces.

The decontamination factors varied from 2.0 to 196.3 for ^{137}Cs and from 1.0 to 30.1 for ^{60}Co . The largest variation between the two isotopes was observed for the NR sample, which is an epoxy-based laboratory coating. Most of the decontaminated isotopes were removed independently from applied decontamination solutions, while the decontamination of CV-coated (strongly rough alkyd resin) samples seemed unaffected by treatment using any of the solutions. The decontamination of other surface materials was less

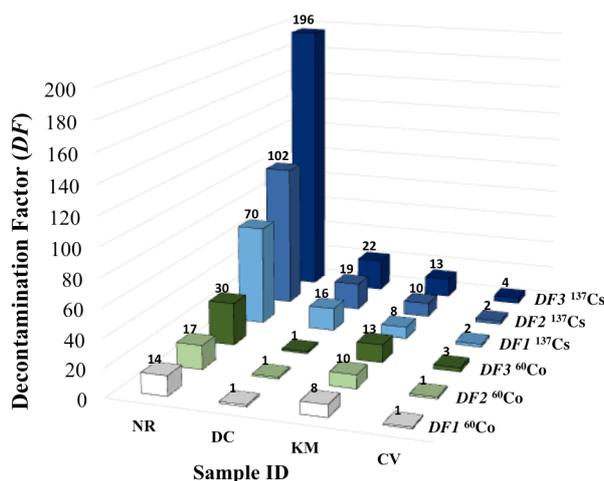


Figure 4. Decontamination factors for MSZ-05 22.7662-83 (Table 2) after drying.

effective than that of the NRs. In the case of KM (glossy enamel paint), the decontamination factor was fair for both isotopes. The results for all materials suggest that decontamination efficiency may be independent of the number of attempts, but dependent on the characteristics of the contamination and features of the surface and contaminant media. In all cases, it seemed that the HCl solution improved the efficiency of decontamination for both ¹³⁷Cs and ⁶⁰Co. Nevertheless, it is notable that distilled water is also a good nominated agent for the decontamination of less specific radioactive cleaning agents.

Table 3 presents the assessment of ease of decontamination using 0.1 cm³ of contamination solution. The ease of decontamination was found to vary from poor to fair for DC, KM and CV. An acceptable level of efficiency was found for NR.

Chemical adsorption was investigated by another decontamination experiment using 0.565 cm³ of contamination solution. Similar results (Fig.5) were observed when compared with the drying method (Fig.4). The decontamination factor using distilled water was found to be approximately the same as for the applied cocktail solution. Similar phenomena were reported by Ruhman *et al.* [1]. The reason for unacceptable efficiencies in the case of samples with CV coatings can be explained by the roughness of the surface, which allows contamination of inner pores due to diffusion, hence the surface becomes very difficult to clean. Surface degradation due to the porosity of the material or by some unknown chemical modification might be another reason. A recommended area for further study is the ease of decontamination in the light of surface changes after years of use [1].

The ease of decontamination was found to vary from poor for CV to fair for DC and KM as shown in Table 4. An acceptable efficacy was found for NR. The ease of decontamination varied from excellent for KM to good for DC and NR. A poor/bad level was found for CV.

Table 3. Assessment of ease of decontamination using 0.1 cm³ of contaminated solution.

samples	isotopes	DF	degree of ease
DC	¹³⁷ Cs	22.5	fair
	⁶⁰ Co	1.5	poor/bad
KM	¹³⁷ Cs	13.4	fair
	⁶⁰ Co	12.6	fair
CV	¹³⁷ Cs	4.0	poor/bad
	⁶⁰ Co	2.7	poor/bad
NR	¹³⁷ Cs	196.3	good
	⁶⁰ Co	30.1	fair

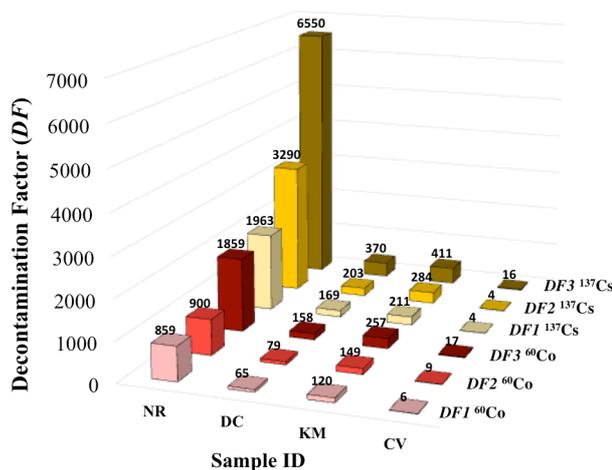


Figure 5. Decontamination factors for MSZ-05 22.7662-83 using the adsorption method (Table 2).

3.2. Decontamination Factors from the ISO Standard 8690:1988

The decontamination factors using the ISO standard method are illustrated in Fig.6. The values clearly show that in the case of epoxy-based NR resin the ease of decontamination was efficient. In the case of the KM coating, the efficiency was the highest. The worst decontamination capability was found for CV-coated samples. The results using the ISO standard clearly show that the ease of decontamination greatly depends on the types of coating. Furthermore, the ¹³⁷Cs isotope can be removed more easily than the ⁶⁰Co isotope, which can be explained by the different physical/chemical properties of investigated isotopes. The obtained decontamination factors for each isotope are summarised in Table 5.

Depending on available conditions, the task and specific conditions, the Hungarian standard method and the ISO standard will be chosen, while the Russian standard was studied only for the sake of comparison. It is important to mention that simulation exercises, for both major and minor contamination events, may be essential for coordination and execution of a response [6–12].

Table 4. Assessment of ease of decontamination in the case of the adsorption method.

samples	isotopes	DF	degree of ease
DC	^{137}Cs	370	fair
	^{60}Co	158	fair
KM	^{137}Cs	411	fair
	^{60}Co	257	fair
CV	^{137}Cs	16	poor/bad
	^{60}Co	17	poor/bad
NR	^{137}Cs	6550	excellent
	^{60}Co	1859	good

Table 5. Assessment of ease of decontamination for the ISO standard 8690:1988.

sample	isotopes	DF	degree of ease
DC	^{137}Cs	684	good
	^{60}Co	315	good
KM	^{137}Cs	4928	excellent
	^{60}Co	3444	excellent
CV	^{137}Cs	13	poor/bad
	^{60}Co	30	poor/bad
NR	^{137}Cs	934	excellent
	^{60}Co	3822	good

3.3. Comparison of DF Values from the ISO and Hungarian Standards

Although a direct comparison of the results between the Hungarian and the ISO standards has limited usefulness and relevance due to fundamental differences in the methods used, we found the same experimental phenomena in terms of ease of decontamination. As expected, similar results were observed and measured for higher decontamination factors in all cases. The most different finding, in the case of KM was that the most effective method observed was by treatment using distilled water. Most of the ^{137}Cs contamination was removed more effectively than ^{60}Co contamination for DC, KM, and NR surfaces.

In comparison with the Hungarian standard method, the ISO standard was used for testing and assessing the ease of decontamination only for chemically adsorbed contaminating isotopes. The experiment was then conducted using 20 contaminated samples of the same type, distilled water as a cleaning agent, and using different decontamination methods. The results are shown in Figs.6-7 and Table 5.

These results are in contrast to our previous observations using the Hungarian standard method and the low efficiency for CV sample. The differences can be explained by some unknown chemical bond formation between the surface of the material and the studied isotopes. This research confirms that the use of deionised water or cleaning agents described in standards may be a sufficient means of removing wet ^{137}Cs and ^{60}Co contamination when identified early.

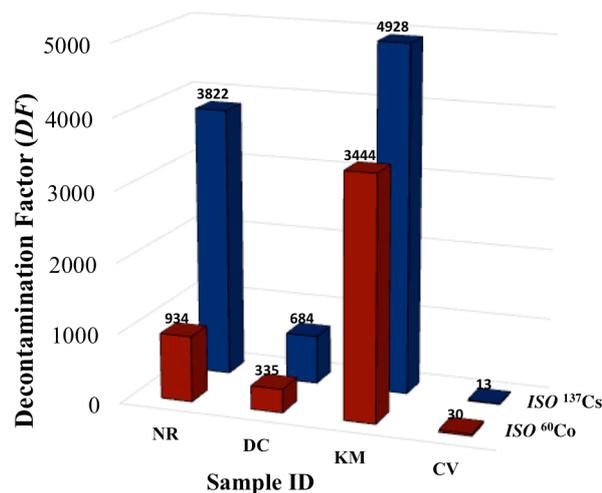


Figure 6. Decontamination factors for the ISO standard 8690:1988 (Table 2).

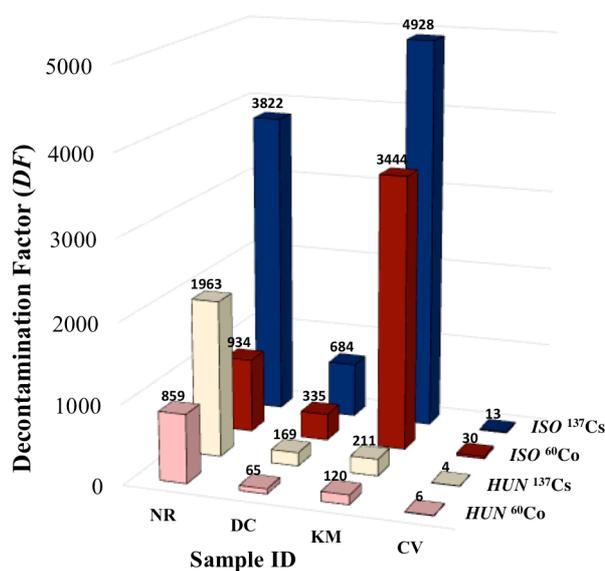


Figure 7. Decontamination factors for the ISO 8690:1988 (E) and Hungarian (adsorption-type contamination) standards.

4. Conclusion

The aim of this study was to identify some of the best surfaces, which would meet ALARA and good-manufacturing-practice requirements to set up protocols to manage contamination in laboratories. The Russian standard can provide very specific information about the ease of decontamination for a wide variety of contamination conditions. The fixed measurement parameters in the case of the ISO 8690:1988 and Hungarian MSZ-05 22.7662-83 standards provide an opportunity to compare paints and coatings and classify them. Using the Hungarian and ISO standards, the ^{137}Cs and ^{60}Co contamination on a surface can be cleaned quickly and effectively using distilled water. Based on the results obtained after the decontamination procedure for the Hungarian and ISO standards, NR can be applied to the surface, the walls of the laboratory, and where

radiation is susceptible in nuclear power plants. In the worst case scenario, when there is an accident involving radioactive contamination, the ability of cleansing the decontamination of surfaces covered by NR will be the most efficient. According to our findings, we were able to select the best materials for the floor of our laboratory.

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