A method and procedure for clearance of slightly contaminated metal materials from Nuclear Power Plant based on the regulatory framework in China

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ABSTRACT

Amounts of slightly contaminated metal with radioactivity were produced during the operation of the nuclear power plant, such as the frame of the waste filter from the ventilation system of nuclear power plants. This created a realistic demand to clear them to implement the strategy of radioactive waste minimization. In this paper, a clearance procedure for the kind of metal materials was established based on the regulatory framework in China. The characteristics of radioactive contamination showed the monitoring of surface dose rate and β contamination could be used as sequential screening methods. Then a detailed radioactivity analysis could be used as a supplementary. This could simplify the procedures and increase efficiency for clearance of the slightly contaminated mental materials. It could also provide a technical base for the radioactive waste minimization in nuclear power plants.

Keywords: Slightly contaminated materials, screening method, clearance level, metal frame of ventilation filter

1. INTRODUCTION

In China, basic laws and regulations have set requirements to reduce radioactive waste from nuclear power plants. Effective methods of minimizing the radioactive waste include the clearance of waste that is exempted from regulatory control[1-5]. In IAEA RS-G-1.7, clearance is defined as the removal of radioactive materials within authorized practices from any further regulatory control by the regulatory body[6]. The Chinese standard GB 18871-2002 provides the clearance rules. It is addressed that the regulatory body shall approve which sources, including materials and objects, within notified or authorized practices may be cleared from regulatory control. The basis for such approval will be the criteria for clearance specified or any clearance levels specified by the regulatory body based on the standard[7].

To clear radioactive contaminated materials, a corresponding radioactivity analysis must be carried out to determine the radioactivity level and determine whether the standards for clearance are met[8]. GB 18871-2002 stipulates the requirement for clearance based on public annual dose limits (≤10 μSv) from practices or sources[7]. As the basic standard for radiation protection, GB 18871-2002 provides exemption guideline levels in an appendix, including total nuclide activities and activity concentrations. It is suitable for the clearance of small batches (usually less than 1 ton) of radioactive materials. However, the framework of GB 18871-2002 considers clearance of small batches of waste unsuitable for radioactive waste generated from nuclear power plants[9]. GB 18871-2002 also provides requirements for the control of surface-contaminated waste. If the surface contamination value is lower than the corresponding limits, it can be considered normal.

The national standard GB 27742-2011 stipulates the clearance level of radionuclides in radioactive waste. It uses the measurable unit Bq/g[10]. The standard applies to activities such as the operation, trade, landfilling, or recycling of large quantities of materials. These are usually larger than 1 ton. The standard also clarifies the clearance level of surface contaminants as the same as GB 18871-2002.

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GB/T 17567-2009 clarifies the requirements for surface contamination monitoring. This applies to recycling and reusing steel, aluminum, nickel, and copper[11]. For the clearance of bulk contaminated materials, a detailed radionuclide analysis is required. The clearance level for surface contamination is consistent with GB 18871-2002.

Based on the consideration of rays released by the decay of radionuclides, most radionuclides will emit X or γ rays at the same time when they decay. The surface radiation dose rate can also be used as an effective measurement for clearance. GB/T 17947-2008 specifies the requirements for measuring radioactive activity for clearance. It is an equivalent standard of ISO 11932-1996[12,13]. Proposed measurement methods include surface α/β contamination, specific activity, surface dose rate, and total activity. The standard proposes that the surface dose rate limit for clearance should take into account the surface dose rate as an excess ranging from 0.05 to 1 μGy/h. This range could apply to both surface contamination and bulk contamination.

Figure 1 shows that China has three levels of radioactive waste clearance as a regulatory framework. The first level is the public exposure dose. The second level is the concentration of radionuclides in the materials. The third level is based on analyzing practices such as surface α/β contamination and surface dose rate measurements as screening methods. In this paper, the indicators system and the analyzing procedure for the clearance level were established in the regulatory framework of China based on the practice of clearance of ventilation filter metal frames, a common metal waste with slight contamination from Chinese nuclear power plants. The suitability of detection and the assessment method for clearance was studied. It could provide a technical basis for the clearing and minimizing of radioactive waste from nuclear power plants.

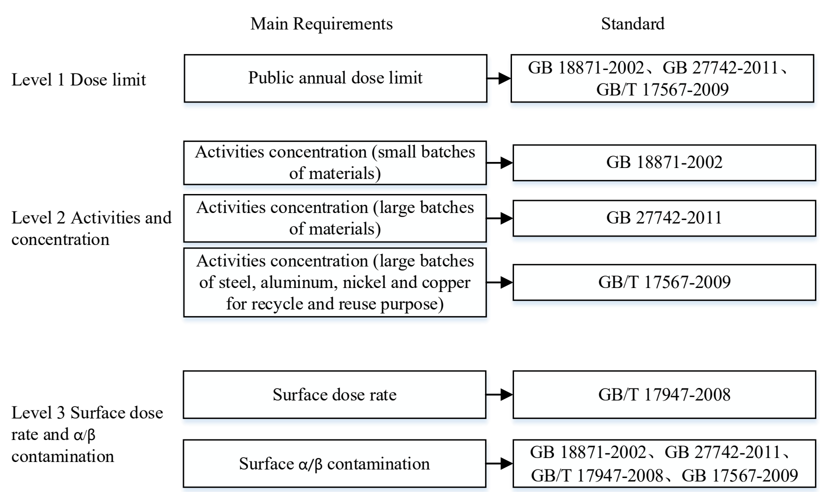

Figure 1. The regulatory framework for clearance in China.

2. CLEARANCE LEVELS AND MONITORING METHODS

2.1 Effective dose levels for clearance

GB 18871-2002 specified the general criteria for exemption or clearance: (1) The effective dose expected to be incurred by any member of the public due to the exempted practice or source is about 10 μSv or less in a year. (2) Either the collective effective dose committed by one year of performance of the practice is about 1 man Sv or less, or an assessment for the optimization of protection shows that exemption is the best choice. (3) The practices and sources are inherently safe. The criteria for clearance are consistent with the requirements of IAEA RS-G-1.7[6].

2.2 Clearance levels

2.2.1 Clearance levels based on the bulk activities. (1) The requirement of GB 27742-2011

GB 27742-2011 specified the activity concentration levels for clearance of different radionuclides[10]. For materials containing multiple artificial radionuclides, the following equation should be met:

$$\sum_{i=1}^{n} C_i / C_{0i} \le 1 \tag{1}$$

where, C_i is the activity concentration of the *i*-th artificial radionuclide in the material, Bq/g; C_{0i} is the clearance concentration of the *i*-th artificial radionuclide, Bq/g; *n* is the number of artificial radionuclides present in the material.

This requirement is the same as that of IAEA Safety Guide No. 44[14].

(2) The requirement of GB/T 17567-2009

GB/T 17567-2009 provides possible clearance levels with 14 radionuclides, such as ^{54}Mn, ^{55}Fe, ^{60}Co, ^{63}Ni, ^{65}Zn, ^{90}Sr, and ^{54}Nb for waste steel, aluminum, nickel, and copper[11]. The clearance levels of these 14 radionuclides were proposed respectively. The calculation models and parameters for these clearance levels were provided. Materials confirmed to be contaminated by these radionuclide contaminants should also meet the clearance levels required by Equation (1). Table 1 shows the clearance levels of these nuclides.

Table 1. Clearance levels for recycling and reuse of radioactive contaminated steel.

Nuclides	Clearance levels (Bq/g)	Nuclides	Clearance levels (Bq/g)
^{54}Mn	4×10^{-1}	^{99}Tc	2×10^{3}
^{55}Fe	1×10^{4}	^{137}Cs	5×10^{-3}
^{60}Co	1×10^{-1}	^{152}Eu	4×10^{-1}
^{63}Ni	1×10^{4}	^{239}Pu	3×10^{-1}
^{65}Zn	6×10^{-1}	^{241}Pu	1×10^{1}
^{90}Sr	9×10^{1}	^{241}Am	3×10^{-1}
^{94}Nb	2×10^{-1}	^{238}U	4×10^{0}

For the bulk contaminated metal frame of the ventilation filter, which is made of stainless steel. According to GB/T 17567-2009, 14 specified radionuclide concentrations should be considered[11]. It is noted that the clearance levels of the 14 nuclides specified in GB/T 17567-2009 are somewhat different from the requirements of IAEA SRS Report No. 44 and GB 27742-2011 (as listed in Table 2). The clearance levels for scrap steel given in GB/T 17567-2009 are slightly higher than those given in GB 27742-2011. It was suggested that the clearance levels of metals should meet the requirement of GB/T 17567-2009 for the 14 radionuclides. Radionuclides not listed should meet the requirement of GB 27742-2011. The clearance level given in GB 18871-2002 is significantly higher than that in GB 27742-2011. Therefore, the clearance for bulk contamination in slightly polluted metal should meet the requirements in the sequence of GB/T 17567-2009, GB 27742-2001, and GB 18871-2002 in China's regulations.

Table 2. Comparison of clearance levels for typical artificial nuclides regulated in different standards (in units of Bq/g).

Nuclides	GB/T 17567-2009 (Steel)	GB 27742-2011	GB 18871-2002	IAEA SRS 44
^{54}Mn	4×10^{-1}	-	1×10^{1}	1×10^{-1}
^{55}Fe	1×10^{4}	1×10^{3}	1×10^{4}	1×10^{3}
^{60}Co	1×10^{-1}	1×10^{-1}	1×10^{1}	1×10^{-1}
^{63}Ni	1×10^{4}	1×10^{2}	1×10^{5}	1×10^{2}
^{65}Zn	6×10^{-1}	1×10^{-1}	1×10^{1}	1×10^{-1}
^{90}Sr	9×10^{1}	1×10^{0}	1×10^{2}	1×10^{0}
^{94}Nb	2×10^{-1}	-	1×10^{1}	1×10^{-1}
^{99}Tc	2×10^{3}	1×10^{0}	1×10^{4}	1×10^{0}

Nuclides	GB/T 17567-2009 (Steel)	GB 27742-2011	GB 18871-2002	IAEA SRS 44
^{137}Cs	5×10^{-1}	1×10^{-1}	1×10^{1}	1×10^{-1}
^{152}Eu	4×10^{-1}	1×10^{-1}	1×10^{1}	1×10^{-1}
^{239}Pu	3×10^{-1}	1×10^{-1}	1×10^{0}	1×10^{-1}
^{241}Pu	1×10^{1}	1×10^{1}	1×10^{2}	1×10^{1}
^{241}Am	3×10^{-1}	1×10^{-1}	1×10^{0}	1×10^{-1}
^{238}U	4×10^{0}	-	4×10^{0}	-

2.2.2 Clearance level based on surface dose rate. GB/T 17947-2008 proposes a dose rate limit for clearance. The excess dose rate near the surface should be in the range of 0.05 to 1 μGy/h[12]. During monitoring, set an alarm limit based on the excess found through surface scanning. If an alarm occurs, the material may have a hot spot for the radioactivity. The standard stipulates the requirements for determining the workplace background[12]. This task in an area is carried out with the lowest possible background. Background measurement needs to be conducted on the site before clearance monitoring. It is noted that the range of 0.05~1 μGy/h is fairly wide, and necessary evaluation should be made to meet the requirement for the clearance determination.

2.2.3 Clearance level based on surface contaminant. GB 18871-2002 provides the pollution control levels of certain equipment and supplies. The regulation applies when their contamination levels fall below 0.8 Bq/cm^2, or 1/50th of the control levels (40 Bq/cm^2), they would be subject to regulatory review as ordinary items[7]. GB/T 17567-2009 and GB 27742-2011 also state that surface contamination clearance levels must meet GB 18871-2002[11].

The main use occasion of ventilation filters in nuclear power plants is ventilation filtration systems. The source term characteristics are consistent with the corresponding gaseous effluent source terms, which are generally β radionuclides. Therefore, an index of surface β contamination of 0.8 Bq/cm^2 specified in GB 18871-2002 should be considered.

2.3 Source terms and the analyzing method

2.3.1 Source terms. Typical ventilation filters used in nuclear power plants include pre-filter, HEPA (High-Efficiency Particulate Air) filter, and iodine trap (Figure 2)[15]. Pre-filter is used to remove a lot of the larger-sized particles so they don't end up clogging HEPA filters. HEPA air filter is effective at capturing almost every size of particle. The iodine trap is used to eliminate iodine dispersion. The source terms for the metal frame of ventilation filters are related to radioactive particles and iodine. For those used in the reactor core area, a possible activation reaction was expected. Radionuclides in the metal frame of ventilation filters may come from neutron activation. Radionuclides may also infiltrate the frame. A surface or bulk contamination was expected. The iodine traps were usually stored for more than years. Therefore, any possible radioiodine would be expected to be nonexistent, due to its short life.

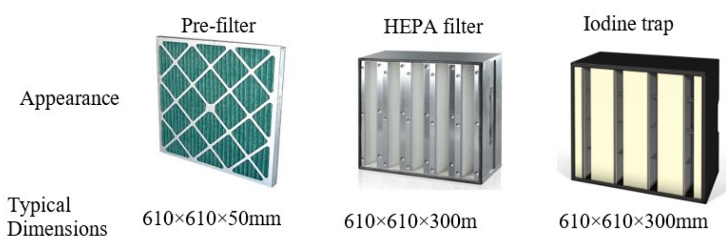

Figure 2. The appearance and typical dimension of ventilation filters in nuclear power plants.

GB/T 17567-2009 addresses 14 radionuclides. Of these, ^{239}Pu, ^{241}Am, ^{241}Pu, and ^{238}U are alpha radionuclides[11], which are unlikely to enter the ventilation system during normal operation of the nuclear power plant. The source term characteristics for ventilation system can be considered when determining radionuclides for clearance assessment. Taking the VVER unit as an example, Table 3 shows the source terms of these nuclides from gaseous effluent[16]. It can be seen that the main source nuclides in the ventilation system are ^{54}Mn, ^{60}Co, ^{90}Sr, and ^{137}Cs, while the annual release amounts of ^{137}Cs and ^{60}Co are significantly higher than the other two nuclides. Therefore, ^{137}Cs and ^{60}Co should be considered as key radionuclides. Metal materials, especially stainless steel, may adsorb ^{3}H (HTO)[17,18]. If necessary, consider taking representative samples for ^{3}H analysis for clearance purposes.

The source term related to the gaseous effluent is related to the possible surface contamination. Beyond that, bulk contamination is expected in the metal frame that operated in the activated area in the reactor. Typical activated products such as ^{54}Mn, ^{60}Co, ^{55}Fe, and ^{63}Ni were expected as bulk contamination. These radionuclides could be considered for analysis with complex separation processes for representative samples from the activated area in the reactor.

Table 3. Airborne effluent emissions of a single VVER unit during normal operation (in units of GBq/a).

Nuclides	Reactor building ventilation system	Auxiliary factory ventilation	Total
^{54}Mn	2.2E-06	6.6E-06	8.7E-06
^{60}Co	2.5E-05	7.6E-05	1E-04
^{90}Sr	5.5E-07	1.5E-06	2.1E-06
^{137}Cs	1.5E-02	4.4E-02	5.9E-02

2.3.2 Analyzing methods. (1) Surface dose rate

According to GB/T 17947-2008 requirements, the measuring range of the portable gamma dose rate meter should be in 0.05~1 μGy/h[12]. During measurement, the net dose rate is obtained by subtracting the background from the scanning values for the surface of the clearance objects. A typical instrument is the FH40G+FHZ672E-10 (Thermo Scientific) radiation dose rate meter. The probe uses a high-sensitivity plastic scintillator with a size of φ 90×90 mm. The measurement range is 1 nSv/h~100 μSv/h, and the energy range is 48 keV~6 MeV[19].

Before carrying out the surface dose rate monitoring, the background level around the workplaces is determined. All workplaces in an area with as low a background as possible are selected. A preliminary investigation of the background radiation levels at the site is required.

After removing the filter material, the frame is cut into regular pieces. They are stacked in groups. Then, the dose rate 5-30 cm from the surface is scanned. The excess dose rate is set as an alarm and the values with the maximum value are recorded. When alarms occur during scanning, the group should be handled for further analysis.

(2) Surface contamination

GB/T 17947-2008 provides the requirements for surface contaminant monitoring. It states that the instrument performance should comply with the requirements of GB/T 5202-2008[12,19]. Typical instruments include the LB 124 monitor (Berthold, Bad Wildbad, Germany) with a large-area ZnS (Ag) scintillator, and the sensitive area is 171 cm^2. The measurement range for the α channel is 0 to 5000 cps. For the β channel, the range is 0 to 50000 cps. The background measurement for the α channel is about 0.1 cps, and for the β channel, it's about 10 cps.

Before surface scanning, the pieces of the metal frame should be rolled out on the workplace. During the canning, the detector should be set as close as possible to the surface. Once the contaminated area is suspected, position the detector and allow it to remain in the same place for about 10 seconds to confirm the value. The scanning speed should match the detector's performance characteristics. For the groups of metal frames without hot spots, only record their maximum scanning value.

(3) γ radionuclides analyzing

A high-purity gamma spectrometer (HPGe) can be used to monitor the activity of γ radionuclides in materials. Passive efficiency calibration software can be used to measure radionuclides in metal sheets when necessary[20].

For the clearance of the metal frame of the waste ventilation filter, ^{137}Cs and ^{60}Co could be used as key radionuclides. Both nuclides are gamma nuclides and can be analyzed on an HPGe. According to a simulation with Ortec Angel calibration software, assuming that a φ75 mm×10 mm metal disc sample is used and measured on an HPGe with a relative efficiency of 50 % (counting time 24 h), the lower detection limit could be approximately 0.001 Bq/ g, significantly lower than the clearance level of ^{137}Cs and ^{60}Co, 0.1 Bq/g. Table 4 shows the comparison of the minimum detectable activity concentration (MDC) for major γ nuclides and their clearance levels.

Table 4. Detection lower limit level of gamma nuclide analysis and the clearance levels.

Nuclides	Gamma energy(keV)	Branching ratio	BG (s^{-1})	Detection efficiency	MDC (Bq/g)	CLs (Bq/g)
^{54}Mn	834.84	0.9997	5E-3	0.004	$<1\times10^{-3}$	1×10^{-1}
^{60}Co	1173.23	0.9985	5E-3	0.0035	$<1\times10^{-3}$	1×10^{-1}
^{65}Zn	1116	0.507	5E-3	0.0035	$<2\times10^{-3}$	1×10^{-1}
^{94}Nb	871.1	1.000	5E-3	0.004	$<1\times10^{-3}$	1×10^{-1}
^{137}Cs	661.66	0.850	5E-3	0.005	$<1\times10^{-3}$	1×10^{-1}

Note: The absorption thickness of iron. The density is 7-8 g/cm^3. The weight of a 1 cm thick sheet is about 300 g. BG, Background; MDC, Minimum detection concentration; CLs, Clearance Levels.

(4) ^3H and other β radionuclides analyzing

To clear the metal frame of the waste ventilation filter, test for the activity of ^3H and other β radionuclides. Then, a complex chemical process is used to extract them from the metal frame. About 50 g of metal pieces could be selected for ^3H extraction. This would be done in a high-temperature oxidation furnace, using tritium-free water as the blubbing liquid for the combustion water collection. The extracting of other β radionuclides such as ^{55}Fe and ^{63}Ni could be performed by digesting with strong acid and then treated as liquid samples according to the related analyzing standards[12,21]. ^3H and other β radionuclides would be analyzed using a liquid scintillation counter (LSC) with the required final samples that were mixed with the liquid scintillator. A typical LSC is the Quantulus 1220 (Pekin-Elmer, Inc., USA). The results should be much greater than the lower detection limit of the LSC as 1 Bq/L for ^3H analysis[22,23]. The feasibility of monitoring is estimated based on one percent of the ^3H clearance level of 100 Bq/g.

3 THE PROCEDURES FOR CLEARANCE MONITORING AND ASSESSMENT

The monitoring process designed to clear the waste metal frame of the ventilation filter is shown in Figure 3.

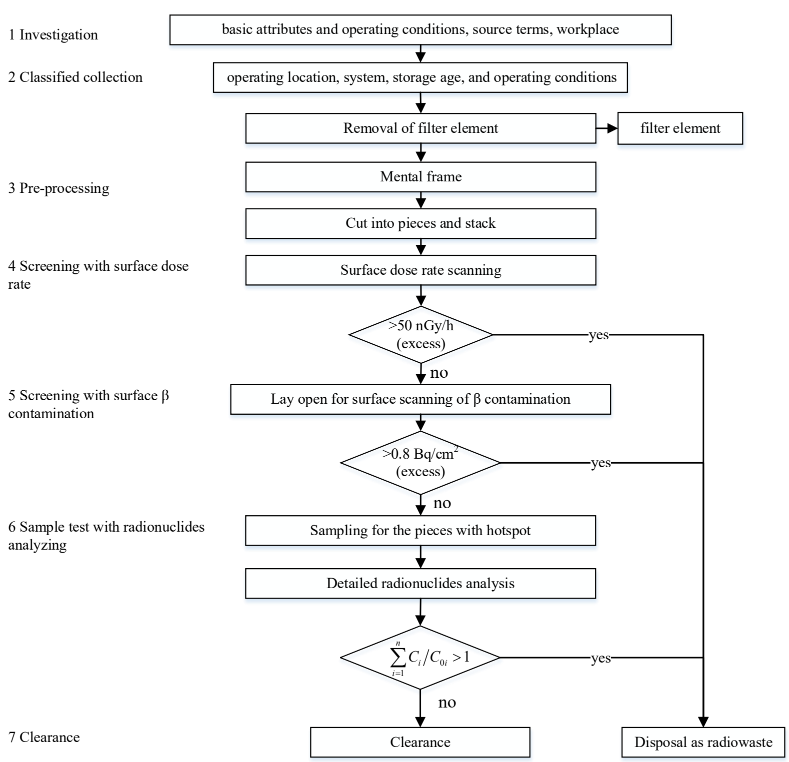

Figure 3. The procedures of clearance and assessment for ventilation filter metal frame.

3.1 Investigation

The contents of the investigation include waste filter investigation, source term investigation, and on-site workplace investigation.

The filter investigation covers the filters' basic attributes and operating conditions. It aims to evaluate the workload and the kinds of possible contamination. It also covers the issues to note during clearance monitoring, etc. It provides an overview of possible contamination of the metal frame of waste filters.

The source term investigation's main purpose is to find the radionuclides that may be contaminated. It also finds the distribution and degree of contamination for the metal frame. The results of source term analysis will eventually be used to select clearance levels and monitoring methods.

The investigation aims to find a suitable on-site workplace. The workplace must have the "lowest possible background radiation" required by GB/T 17949-2008[12]. The investigation also seeks a suitable area for dismantling, decontamination, and radiation monitoring.

3.2 Classified collection

Classified collection helps distinguish the sources of waste filters. It lets targeted clearance monitoring be done. Based on the investigation of the filter, source term, and workplace, the classification and collection of waste filters and the division of placement areas were carried out at the controlled workplace. The classification method can be classified according to factors such as the operating location, system, storage age, and operating conditions of waste filters. The filters' operating area or system should get preliminary consideration. This is from the perspective of possible neutron activation. Consider the core activation area and general areas.

3.3 Pre-processing

Pre-processing waste filters mainly means disassembling them and removing their filter elements. The elements include high-efficiency and primary filters made of glass fiber and activated carbon. It also involves cleaning the filter's surface and disassembling its metal frames. They are rolled out and cut into pieces to be suitable for stacking or surface monitoring. The filter and activated carbon go in one barrel. The cotton waste from cleaning goes into another.

3.4 Screening with surface dose rate

Surface dose rate monitoring is a rapid screening method for clearance monitoring, which is suitable for surface contamination and bulk contamination. It could be estimated for the relation of surface dose rate and the surface activity concentration by the Monte Carlo simulation method. The typical source has radionuclides of ^{60}Co and ^{137}Cs. For a proposed iron sheet with dimension about 100 mm×100 mm×2 mm, the surface activity concentration of 0.8 Bq/cm^2 for ^{60}Co and ^{137}Cs would generate a surface dose rate of 178 nGy/h and 55.9 nGy/h at 5 cm distance, respectively, meanwhile, a surface dose rate of 57.5 nGy/h and 15.2 nGy/h at 30 cm distance. So, the conservativeness could be guaranteed with a screening method for surface gamma dose rate in the situation of the main surface source terms of ^{60}Co and ^{137}Cs.

Beyond the simulation of the relation between the surface gamma dose rate and surface β contamination, the relation between the surface gamma dose rate and the bulk contamination were stimulated with Monte Carlo simulation method. A stacked cubic with iron sheets about 100 mm×100 mm×2 mm with the interval about 2 mm were proposed for the simulation. It could be obtained that the bulk contaminated iron with 0.1 Bq/g for ^{60}Co and with 0.5 Bq/g for ^{137}Cs as the clearance levels would generate a surface gamma dose rate of 407 nGy/h and 489.5 nGy/h at 5 cm distance from the up surface of the proposed cubic, respectively. For the distance of 30 cm, the corresponding estimated value are about 232 nGy/h and 236.5 nGy/h.

It could be concluded that the surface dose rate of 50 nGy/h as an excess could be used as the screening for the clearance assessment conservatively. For the waste metal group with surface excess dose rate lower than 50 nGy/h, the materials could be regarded as clearance, while that over 50 nGy/h should be analyzed further with surface β scanning. Much attention should be paid to the materials from the reactor core area, higher surface dose rate is expected.

3.5 Screening with surface β contamination

The Chinese rules regulated surface contamination at 0.8 Bq/cm^2. It could be used as the second screening level for clearing the waste metal frame of a ventilation filter. The detailed operation process and the attention points could be obtained as described above. If the excess surface β contamination was below the limit of 0.8 Bq/cm^2, it was clearance waste. If it was over 0.8 Bq/cm^2, it should be used as a sample test. The test should check for γ radionuclides and ^{55}Fe, ^{63}Ni, ^{3}H, and others. If the scanning approves the possible hot spot, the possible smear or other practical decontamination method could be used, and the further surface scanning with the monitor to check the effect of decontamination could be conducted[24]. If the excess values were still higher than the limit of 0.8 Bq/cm^2, it should be checked with more radionuclides analysis.

3.6 Sample test with radionuclides analyzing

According to the requirements of GB/T 17567-2009[11], in order to prevent the existence of radioactivity hot spots, all measurements should be carried out in accordance with relevant quality assurance requirements and should ensure that they meet the sample representativeness and quantitative statistical requirements. The highest mass activity concentration cannot exceed 10 times the average. The representativeness judgment condition of sampling is $\max(x_i) < 10 \times \bar{x}$.

3.7 Sampling review

After screening and monitoring, determine which materials can or cannot be released. The sampling review is mainly aimed at materials with hot spots detected, especially materials that cannot be released. Further confirmation is required, and some samples are taken for surface contamination monitoring to confirm the level of possible radioactive contamination.

3.8 Clearance

The waste metal frames that are confirmed to be released after clearance monitoring and assessment can be temporarily stored and subsequently recycled and disposed of according to the decision of the power station, while those cannot be released should disposed of as radioactive solid waste.

4. CONCLUSION

A reasonable and effective assessment procedure is critically required for the clearance of radioactive contaminants, and the key point lies in the efficient and accurate methodology for radiation monitoring and radioactivity analysis. In this paper, we established a practical procedure for the clearance of the slightly contaminated metal materials taking the metal frame of ventilation filters from nuclear power plant based on the regulatory requirement in China. A sequential screening procedure for surface dose rate, surface β contamination, and detailed radionuclides analysis was established based on Monte Carlo simulation for the screening levels, which could make the clearance work much more effective.

It should be noted that the current procedure is mainly based on sampling of the radioactive waste. Heavy monitoring work could be expected from the representativeness point when large quantities of materials are needed for clearance. It is necessary to develop an automated monitor to improve efficiencies, simplify work processes, and improve accuracy, which could guarantee that clearance materials will not pose possible radiation risks to the public.

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