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THE FREE RELEASE OF THE MATERIALS RESULTING FROM THE DECOMMISSIONING OF THE VVR-S RESEARCH REACTOR

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Abstract. All materials used during the nuclear reactor operation are potential radioactive wastes. The role of the free release of materials from regulatory control is to reduce the amount of radioactive wastes. The CNCAN allows licensees to release materials according to pre-established criteria which comply with Romanian standards and international recommendations. In this paper, the methodology used for free release of the materials arising from the VVR-S reactor decommissioning is described and commented.

Key words: Free releases; decommissioning; standards and recommendations; release criteria.

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1. INTRODUCTION

The Măgurele (Romania) VVR-S nuclear reactor is a research reactor with a maximum thermal power of 2 MW, and distilled light water as moderator, coolant and reflector. The reactor produced 9.59 GWd during its 40 years of operation. The last shut down was in July, 1997. In 2002, Romanian Government Decision decided to shut down the reactor for subsequent decommissioning. Decommissioning activities started in 2010, and are planned for an 11-years period.

Before decommissioning start, the reactor was kept in conservation phase. During this phase, under BOA 3J -00 201 Basic Ordering Agreement Project, concluded between IFIN-HH and U.S. Argonne National Laboratory, clean-up and radiological characterization were performed. As a result of the clean-up activities, a significant amount of materials was released from regulatory control, *i.e.* about 44.8 tons collected from the controlled area and 64.8 tons from supervised area. Further, the information provided by the radiological characterization [1], was use to classify the materials to be released.

In reference [2], it is mentioned the radiological inventory estimation for the nuclear reactor block and the amount of radioactive waste and low activity materials which could to be released from regulatory control. These study results allowed us to appreciate that more than 80 % of the reactor block materials can be released.

2. INTERNATIONAL RECOMMENDATIONS

It is worth mentioning that all international organizations provide special care to free release of materials from regulatory control by following a set of recommendations and guidelines, based on radiological risks, and special dedicated to this process.

2.1. INTERNATIONAL ATOMIC ENERGY AGENCY RECOMMENDATIONS

Safety standards [4,5] and supporting documents [6,7], published by IAEA are a set of regulatory-style publications that give an international consensus on the radiation protection principles. According to the radiation protection and safety basic principles, which were developed by the International Commission on Radiological Protection (ICRP) and the International Nuclear Safety Advisory Group (INSAG), IAEA introduced and defined, in 1996, the concepts of i.- exclusion, ii.- exemption and iii.- clearance which could be found in International Basic Safety Standards (BBS) [4].

i. - exclusion concept is related with the ionizing radiation excluded from Regulatory Control because the control is not possible. Excluded exposure examples, given in the BSS, are: exposure from ^{40}K in the body, from cosmic radiation at the surface of the earth and from unmodified concentrations of radionuclides in most raw materials. Such sources are excluded from control by their nature. With regard to radiation sources, these shall be controlled before returning to the public by the following routes: exemptions or clearance.

ii. - exemption concept has concern with practices and sources within a practice which can be, *a priori*, exempted from standard requirements (not from legal framework), if exposures or risks shall be sufficiently small.

iii. - clearance concept is concerned with the removal of radioactive materials or radioactive objects within authorized practices from any further regulatory control. Various terms are used in different States to describe the clearance concept, *e.g.* free release [8].

IAEA published in 2004 the Safety Standards Series No. RS-G-1.7 [5],whose objective is to guide the national authorities on the application of the concepts of exclusion, exemption and clearance. Section 4 of RS-G-1.7 provides activity concentration levels (Bq/g) for exclusion, exemption and clearance. These levels were determined assuming that the exposures would be of the order of 10 μSv within a year. The methodology and parameters used to calculate these activity concentration levels were provided by the IAEA in the [6].

Over the last decade, IAEA Member States have made considerable efforts to develop consensus on application of the exclusion, exemption and clearance concept. In January this year, as response to the Member States requirement, IAEA provided in Safety Reports Series no. 67 [7], practical examples on the application of the exemption and clearance levels and the activities required for the demonstration of compliance with these levels.

2.2. EUROPEAN UNION STANDARDS

In the European Union (EU), national legislation on radiation protection is bound by the EURATOM Treaty to comply with the EUs general Basic safety standards [9]. This standard contains general provisions for clearance. European Commission has set levels of clearance and published them in the Series Radiation Protection (RP): clearance levels for metals RP 89 [10], clearance of buildings and building rubble RP 113 [11], unconditional clearance of all types of materials: RP 122 part I [12].

In 2010, the European Commission published the RP 157 [13] study that compares the guidelines recommended by the IAEA and EC on exemption and clearance levels. The comparison presented in this study is made between RP 122 [12] and RS-G-1.7 [5]. The RP 157 study provide values of the ratio between levels given in RP 122 and RS-G-1.7 and explanations for differences between calculated values of levels in these two documents.

3. ROMANIAN REGULATIONS APPLICABLE TO THE FREE RELEASE

In Romania, the National Commission for Nuclear Activities Control (CNCAN) is the national authority competent in exercising the regulatory activity, authorization and control in the nuclear field, as provided by the Law 111/1996, republished in 2010, regarding the safe deployment of nuclear activities. There are two types of CNCANs norms regulating the free release of the materials arising from authorized nuclear activity: (i) Radiological safety norms (NSR) and (ii) Norms on radioactive waste management (NDR).

Radiological Safety Fundamental Norms [15], which are included in the set of

Table 1.

The exemption and release levels according to CNCAN NSR-01 [15].

Exemption levels	Values established by CNCAN, expressed in terms of total and specific activity, below which the practice is exempted from the authorization requirements of NDR-02.
Release levels	Values, established by CNCAN, and expressed in the terms of activity concentration and surface activity, below which radioactive materials, regardless practice and subject to the authorization requirements, can be released from the requirements of NDR-02.

norms dedicated to radiological safety, are based on the Council Directive 96/26/EURATOM [9] and on the IAEA BSS [4] conditions. The levels of exclusion and exemption are given by CNCAN in Annex 2 of these Norms. Its contents are explained in the terms of exemption levels and release levels as illustrated in Table 1.

Norms concerning the release from regulatory control of materials arising from authorized nuclear activities [16] include the set of norms dedicated to radioactive waste management. Objective of these norms is to release the materials resulting from nuclear activities in such a manner, that the radioactivity content of these materials shall not involve a significant risk for public and environment. It is considered that the risk is not significant for public and environment if after the release, as a result of analyzing exposure pathways, it is unlikely that the annual effective dose of any person from the public to exceed 10 μSv and it is practically impossible to exceed 100 μSv.

The release of materials from the regulatory control could be unconditional or conditional. In the first case, the materials can be subsequently used without any restriction. In order to accomplish that, the material must meet the exclusion requirements of the NSR-01 [15] Annex 2 in the case of a single radionuclide. For a mixture of radionuclides, according to the norm NDR-02 [16], the release criterion is:

$$\sum_{i=1}^{n} \frac{\Lambda_i}{RL_i} < 1 \tag{1}$$

where n is the number of radionuclides in the material, Λ_i and RL_i are the specific activity and release level corresponding to the i-th nuclide.

In the case of the conditional release, the materials can be recycled / reused only according to the release conditions. According to the norm NDR-02 Annex 2, the authorization holder must get in advance the Ministry of Health authorization and CNCAN approval for using higher values for release levels than those given by NSR-01 for the respective materials. For release levels compliance verification, according to NDR-02, the following methods of measurement are recommended by CNCAN, as appropriate: (i) direct measurements, (ii) laboratory measurements for representative samples, (iii) use of scaling factors properly derived, (iv) other methods accepted by CNCAN.

4. SELECTION OF MONITORING TECHNIQUES AND INSTRUMENTS USED FOR FREE RELEASE

The selection of monitoring techniques and instruments depend primarily on the radionuclide or radionuclide mixture which has to be measured. As a result, it is important to know the radionuclide inventory of a nuclear reactor. The radionuclide inventory of the VVR-S, Măgurele reactor is due to reactor operation, radioisotope production in hot cells, depleted uranium processing and research activities. Due to the fact that the VVR-S Măgurele reactor was shut down 15 years ago, only radionuclides with half live higher than one year have a significant contribution to the radionuclide inventory. In Table 2, are shown the major possible radioactive contaminants in the VVR-S reactor, their radiations, their half lives and the release levels specified by NDR-02.

As it is shown in Table 2, radioactive contaminants from VVR-S reactor can be measured by gross beta-gamma counting, gross alpha counting and gamma spectrometry. The activity of hard-to-detect radionuclides is difficult to be measured, but it can be correlated with the activity of key radionuclides (nuclides that emit strong gamma rays, *e.g.* ^{60}Co) by means of scaling factors. The values of the scaling factors thus calculated could be found in the Table 3 of Ref. [2].

The radionuclide inventory of VVR-S nuclear reactor falls into two categories: surface contaminated materials and bulk contaminated or neutron activated materials. Three measurement techniques are used for free release of materials: bulk monitoring (especially for gamma emitters), surface monitoring (predominantly for beta and alpha emitters) and radiochemical analysis. The last is often used to determine the nuclide composition of a given object or a waste stream.

4.1. SELECTION OF MONITORING TECHNIQUES AND INSTRUMENTS FOR SURFACE CONTAMINATION IDENTIFICATION AND MEASUREMENT

Surface contamination surveys are performed using both scans and static direct measurements. If necessary, smear samples can also be used for non-fixed contamination indirect measurements. Monitoring of the surface contamination is based on the use of hand-held instruments which should have a Minimum Detectable Activity (MDA) less than the applicable release criterion. An important aspect of release surveys is to adequately assess the background levels associated with specific materials, by using materials that have no reasonable potential to be contaminated. Scanning is

Table 2.

Major possible radioactive contaminants due to the VVR-S reactor operation

Radionuclide	Emitted radiation	Half-life (y)	Release level: Surface activity ($Bqcm^{-2}$)	Release level: Specific activity (Bqg^{-1})	Remarks
Activation products					
^{3}H	β^- (100 %)	12.3	1000	200	hd
^{14}C	β^- (100 %)	5.73 10^3	30	20	–
^{55}Fe	EC, X	2.7	300	30	hd
^{63}Ni	β^-	100	1000	70	hd
^{60}Co	β^-, γ	5.3	3	1	kr
^{134}Cs	β^-, γ	2.4	10	7	–
^{152}Eu	EC, X, β^-, γ	13.5	10	7	–
^{154}Eu	X, β^-, γ	8.6	3	5	–
^{155}Eu	X, β^-, γ	4.76	30	30	–
Fission products					
^{137}Cs	β^-, γ	30	3	0.8	kr
Actinides					
^{138}U	α, γ	5.4 10^9	1	0.02	–
^{241}Am	α	432	0.3	0.05	kr

hd - hard to detect radionuclide; kr - key radionuclide

performed in order to identify areas with elevated activity and determine the degree of homogeneity of contaminated surfaces. Alpha and beta radiation measurements are mainly performed using scintillation detectors, proportional and Geiger-Muller counters. The majority of these detectors are planar detectors with a thin entrance window. The detectors result is expressed in counts per second, which then must be converted to usable physical quantities, generally (Bq/cm^2). According to ISO 7503-1 [17], the surface activity Λ_S of the fixed and non-fixed contamination, in (Bq/cm^2), is given by the following equation

$$\Lambda_S = \frac{R_T - B}{\varepsilon_i \varepsilon_s S}, \tag{2}$$

where R_T is the total count rate, in reciprocal seconds; *B* is the background count rate, in reciprocal seconds; ε_i is the instrument efficiency for beta radiation; ε_i is the efficiency of the contamination source.

In situ, the contamination surfaces efficiency determination is difficult to assess and depends on the type of material and contamination distribution in depth. A new method [18] has been developed for determining the efficiency of the large-area beta sources. This method is based on the measurement of the transmission coefficient of beta particles through a thin aluminum foil, and it was used for determining the efficiency of a $^{90}Sr/^{90}Y$ reference source. On this method basis, we propose to determine the efficiency for different materials types from the VVR-S reactor decommissioning and for the radionuclides of interest.

In some case, where large concrete surfaces or soils have to be monitored to low levels for unrestricted release, gamma spectrometry might be operated. However, regarding the measuring times, this method is not advisable on surfaces area less than 1 m^2, which has to be compared with the averaging area accepted by the national authorities. Gamma gross counting can also be operated, either with plastic scintillators, or with more efficient inorganic scintillator such as NaI(Tl). Because of the poor resolution of NaI(Tl), this type of detector is not suitable for the identification of complex mixtures of gamma emitting radionuclides. Scenarios requiring such analyses involve detectors with higher resolution such as HPGe detectors.

4.2. SELECTION OF MONITORING TECHNIQUES AND INSTRUMENTS FOR NEUTRON ACTIVATED MATERIALS

Various techniques are available for the measurement of specific activity (Bq/g). They are appropriate when either neutron activation has taken place or where contamination has diffused into or become mixed with the material in question. Circumstances may arise when materials were contaminated with alpha and beta radionuclides which have negligible X and gamma emissions. Such radiations (alpha and beta) are difficult to detect in bulk samples. The very limited range of alpha and beta radiations dictates that only the surface or near surface of a sample contributes to the signal. Hence it will be essential that either any measurement sample is homogeneous or sufficient samples are taken from an inhomogeneous material to allow a confident calculation of the specific activity of a bulk sample equal to the permitted averaging mass.

Gamma spectrometry or gross gamma activity measurements are usually performed for measuring the specific activity of activated materials arising from the decommissioning of nuclear reactors. Gamma spectrometry techniques can be divided into two categories: (i) high resolution gamma spectroscopy (HRGS) and (ii) low resolution gamma spectroscopy (LRGS). Gamma spectrometric techniques involve measuring the energy spectrum of the gamma-rays originating in the sample and using specific features within this spectrum to identify and differentiate between

Table 3.

MDA of ISOCART system in the measurement of 200 litre drums.

Nuclide	MDA kg m^{-3}		
	Density (0.3 gcm^{-3})	Density (0.5 gcm^{-3})	Density (1.0 gcm^{-3})
^{60}Co	0.38	0.48	0.74
^{137}Cs	0.56	0.75	1.10

gamma-rays from different isotopes as well as to quantify the amount present. Gross gamma counting simply counts the total number of gamma-rays reaching the detector without heed of their energy (some threshold selection criterion may be applied).

Mobile and hand held instruments with spectrometer capabilities are very useful for *in situ* measurements. They vary from completely self-contained instruments to instruments which acquire and store spectra, and require connection to a personal computer for interpretation.

ISOCART Mobile Assay System is an ideal solution when the measuring instrument has to be frequently moved. It provides all necessary hardware and software, with a choice of detectors and collimators in a highly mobile format. Also, ISOCART spectrometric system for waste assay is versatile due to its ability to measure a wide range of waste containers and different objects. The detection efficiency of the ISOCART system for a given energy depends not only on the energy and experimental set up, but also on the radionuclide. Thereby, a comprehensive experimental calibration would require the measurement of an enlarged number of standards, one for each geometry and matrix of interest, containing certified activities for each radionuclide that is present in the real samples. This task is almost impossible for the majority of the matrices and radionuclides. A better solution for determining the detection efficiency is the application of specific methods of calculation. A computation procedure, based on Monte Carlo simulation of spectrum and detection efficiencies for ISOCART system used currently for the radioactive waste drums measurement, was developed by Radu and Sima [19]. ISOCART system can be used for the purpose of free release of materials from the regulatory control because it has a low Minimum Detectable Activity (MDA). In view of this, Table 3 shows, according to the ORTEC online documentation [20], the typical detection limits in the measurement of 200 litre drums containing ^{60}Co and ^{137}Cs by using a 80 % relative efficiency HPGe detector.

The CCM grind box monitor is an example of gross gamma counting technique based on the utilization of plastic scintillator detectors. It is used for release measurement of grid boxes filled with system components, demolition materials etc. The monitor is based on the CCM (Cobalt Coincidence Method), which is ^{60}Co nuclide specific detection, spatially focusing and even has no cross-sensitivity against interference from ambient dose rate. It consists of four plastic scintillation detectors, $1000\times500\times100$ mm, with integrated photomultiplier and voltage divider. One detector is mounted on each side of the assembly. The efficiency of the detector arrangement, related to 4π geometry (measured under operation conditions, point source in centre position) are 0.4 (s^{-1}/Bq) for integrally or respective 0.003 (s^{-1}/Bq) measured in CCM mode for ^{60}Co radionuclide. Integral background is around 5000 cps/per detector and around 5 cps sum for CCM mode. During measurement, the system shows in the main measuring program window ^{60}Co CCM, ^{60}Co window,

^{137}Cs window and integral activities (in kBq, Bq/cm^2 or Bq/g), as well as the detection limits currently reached in percentage of detection limit values adjusted by the user.

5. THE METHODOLOGY USED FOR FREE RELEASE OF THE MATERIALS ARISING FROM THE DECOMMISSIONING OF THE VVR-S REACTOR

All materials (equipment, components or parts of equipment, tools and devices or any other object) used during the nuclear reactor operation are potential radioactive wastes. The role of the free release of materials from regulatory control is to reduce the amount of radioactive wastes, demonstrating that these materials meet the release criteria.

To demonstrate the compliance with the release levels specified by NDR-02, the free release of materials from the VVR-S reactor is conducted within an appropriate program of quality assurance. The methodology used for free release of contaminated materials arising from the decommissioning of the VVR-S reactor includes three stages: preliminary stage, decision stage and reporting to CNCAN stage. It allows a gradual approach and an iterative release process in order to obtain a high level of confidence that the requirements of the NDR-02 are fulfilled. In the case of neutron activated materials, an average value of the specific activity (Bq/g) over a specified value of the mass is measured using different methods (usually gamma spectrometric methods).

5.1. PRELIMINARY STAGE

Figure 1 presents the steps to be followed in the preliminary stage. The result of this stage is the material placement in one of the three classes of potential contamination or/and activation. According to NUREG 1761 [21], the classification of materials is done as follows:

Materials from Class 1 are those materials that have a high potential for contamination or/and neutron activation and a high probability for exceeding release levels. These materials were in direct contact with radioactive materials or were located in the reactor block and activated.

Materials from Class 2 are those materials that have a small potential for contamination or/and activation, but are not expected to exceed the release levels. These materials include those items that are within controlled area, but are not expected to have contamination.

Materials from Class 3 include those materials that were not activated, and either it is not expected to contain any contamination, or it is expected to contain contamination less than release levels. The most important activities performed in

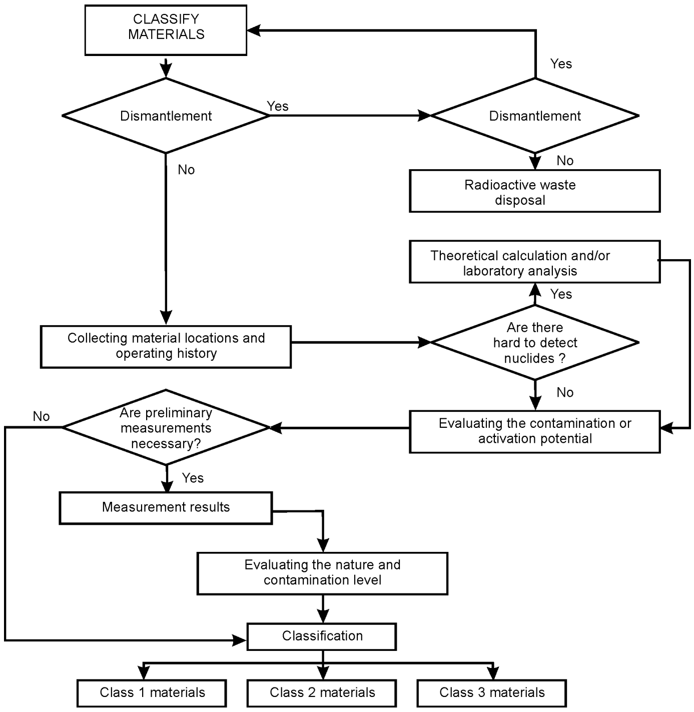

Fig. 1 – Flow-chart illustrating the methodology used for the free release of the materials arising from the VVR-S reactor: preliminary stage.

the preliminary stage are: (i) physical description of materials; (ii) collecting and evaluating information concerning the materials location and operating history; (iii) evaluating and documenting the contamination or activation potential and evaluating the nature of contamination. Based on the information obtained from the activities above mentioned, the classification of materials can be done according to NUREG 1761 [21].

5.2. DECISION STAGE

In the decision stage (Figs. 2 and 3), is presented the selection of measurement techniques depending on the material classification (see Fig. 2). As stated in the preliminary stage classification, this is based on the potential of contamination or activation and on the release levels.

Comparing the results from surface activity (Bq/cm^2) or specific activity (Bq/g) with the release levels specified by norms, each material route is established as follows: (i) For 3 and 2 class materials, which meet the conditions stipulated in the NDR-02 [16] and below the exclusion level of the NSR-01 (2000) Annex 2 [15], unconditional release documentation is prepared. Materials that do not meet these

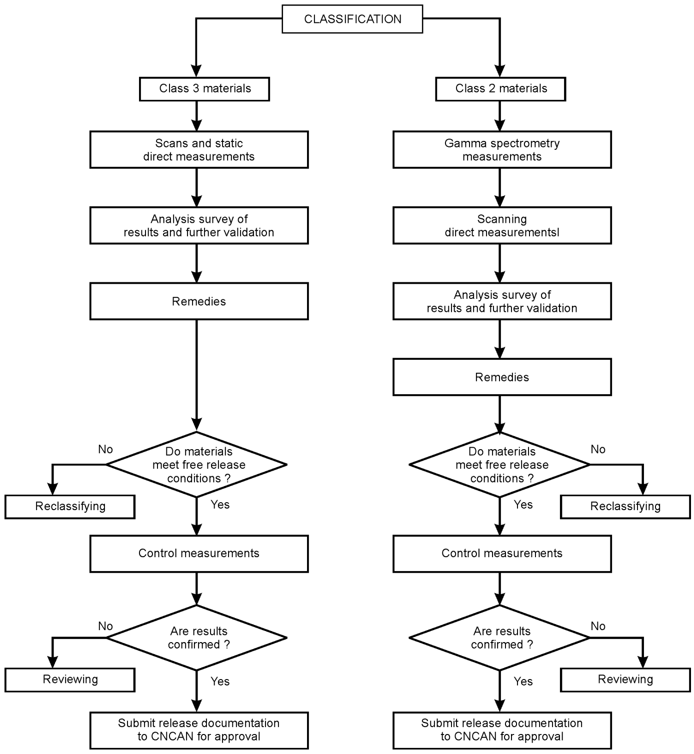

Fig. 2 – Flow-chart illustrating the methodology used for the free release of the materials arising from the VVR-S reactor: decision stage for materials of classes 2 and 3.

conditions are reclassified as Class 1. (ii) For Class 1 material, a technical and economical analysis is prepared, in order to determine whether their decontamination is effective.

For these materials, which are above the level of exclusion, the possibility to be unconditionally released, accordingly to NDR-02 Annex 2, is analysed. Materials that do not meet authorization release requirements shall be treated as radioactive waste. The solid materials that do not meet exclusion requirements of the NSR-01 Annex 2, but meet the provisions of the NDR-02 Annex 2, can be released only if the authorization holder gets in advance the Ministry of Health (MH) authorization and the CNCAN approval for using the obtained values as release levels for the respective materials, according to NDR-02 Annex 2.

6. CONCLUSIONS

The presented methodology regarding free release of the materials arising during the decommissioning of the VVR-S, Măgurele reactor, complies with all Roma-

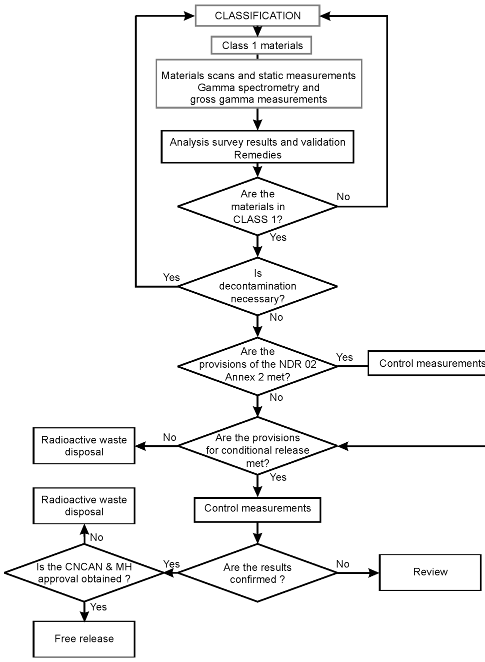

Fig. 3 – Flow-chart illustrating the methodology used for the free release of the materials arising from the VVR-S reactor: decision stage for materials of class 1.

nian standards and international recommendations that regulate the nuclear industry.

In terms of measurement, this strategy meets the CNCAN requirements, and it comprises at least the following six steps: grouping of materials to be measured in order to ensure the radionuclides spectrum homogeneity assessment of radionuclides spectrum for the materials to be released by sample analyses, taking into account all pertinent information about the materials operational history choosing of the suitable measurements method in order to demonstrate the released levels observance choosing of the suitable measurement instruments and their appropriated calibration establishing of the appropriate measurement, recording and reporting.

By applying this methodology, we estimate that about 110 tons of materials could be unconditionally release so far. To this should be add the amount of about 582 tons of materials resulting from the decommissioning and dismantling of the VVR-S Măgurele reactor block.

Therefore, it is of a great importance for the decommissioning process, to demonstrate that the global activity of a large part of these materials is below the norm limits and could be free released from regulatory control.

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