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The Assessment of the Radioactive Inventory for the Solid Wastes from VVR-S Nuclear Research Reactor Decommissioning

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The radioactive inventory of the solid wastes resulted from decommissioning of the VVR-S research reactor from Bucharest, Romania, is assessed and compared to the estimated one. The main part of the inventory is concentrated in the reactor block and hot cells. The reactor block inventory was estimated by preliminary measurements and calculation before decommissioning. The activated and contaminated structures were dismantled, cut, placed in containers and analysed by gamma ray spectrometry method. The dominant radionuclide was ^{60}Co identified in all dismantled components. By comparison of the activities measured in 2017 with those estimated in 2012 it can be noticed a good agreement between the estimations and the actual situation. The radioisotope production generated also a significant inventory at about 10.57 Ci.

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1. Introduction

The VVR-S nuclear research reactor with thermal neutrons from Romania, moderated and cooled with distillate water was the first one of this type put into operation in the South-East of Europe. Used mainly for research and radioisotope production purposes, the reactor was operated without any incident, between 1957÷1997 at a nominal thermal power of 2 MW, using low-enriched (10% ^{235}U) EK-10 type and high-enriched (36% ^{235}U) S-36 type nuclear fuel. The spent fuel was repatriated in Russian Federation. The reactor decommissioning represents the final step in the lifecycle of the nuclear facility, after operation completion and final shutdown. This process started in 2010 and will be finished in 2020. The present paper performs the analysis of the reactor block and hot cells radionuclide inventory during the decommissioning period in comparison with its level from the normal operation period. The decommissioning generated significant amounts of solid wastes which were managed according to the degree of activation/contamination, the type and the half-life of the contained radionuclides. The wastes are conditioned for final or intermediate storage. The activated/contaminated components of the reactor block are cut into pieces whose size allows the safe placement into a container presented in Fig. 1a which can be 220 l carbon-steel container shown in Fig. 1b or 260 l ekolRAD 50 cast iron container shown in Fig. 1c, according to the level of activity, the type of material and the adopted treatment/storage method. Prior to the cutting, the dismantled parts are checked in order to determine the degree of surface contamination and have been decontaminated using the Decon-Gel solution — a polymer that allows the radioactivity confinement. The containers are then measured by gamma-ray spectrometry method at the Radiological Characterisation Laboratory (RCL) of Reactor Decommissioning Department (RDD)

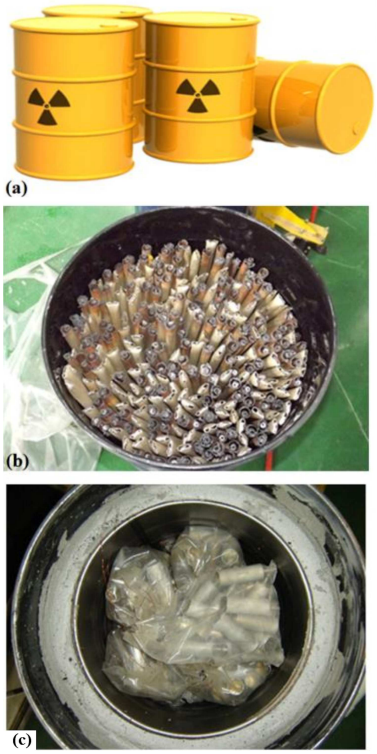

Fig. 1. (a) Containers for radioactive waste storage, (b) carbon steel container, (c) ekolRad container.

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before the intermediate storage or transfer to RWMD (Radioactive Waste Management Department). The aluminium – low intermediate level waste–short live (LILW-SL) type — is stored in ekolRAD 50 containers and the graphite — low intermediate level waste–long live (LILW-LL) type — is stored in the spent nuclear fuel storage pools, due to the fact that a conditioning technology for final storage under radiological safety conditions is not available. All other waste will be permanently stored at the National Radioactive Waste Repository (NWRW).

2. The methodology for the inventory assessment

The paper makes an analysis of the solid wastes inventory resulted from reactor block and hot cells dismantling. Before the reactor decommissioning, the radiological characterization was performed (2004÷2007) in order to determine the activity inventory and contamination level of the structures and systems. The main component of the inventory was generated due to the neutron activation of the building materials (aluminium, cast iron, concrete and graphite) located in/or near the active area of the reactor block (see Fig. 2). The concrete biological shielding was exposed to a low neutron flux. The radioisotopes production generated significant contamination in the reactor building, ventilation system, also in the radioactive liquid waste collection system. According to the paper [1] the main contaminants are: ^{60}Co, ^{134}Cs and ^{137}Cs and ^{238}U (resulted from the depleted uranium processing). The reactor block inventory was estimated also by Ionescu et al. [2] in 2012. The estimation was based on the space distribution of the thermal neutron flux inside the reactor structures and on gamma ray spectrometry results for samples taken out of the reactor core, reactor base plate, thermal column and different places of the biological shielding. For easy-to-measure radionuclides such as: ^{60}Co, ^{152}Eu, ^{154}Eu and ^{155}Eu the specific activity was calculated by means of gamma spectrometry data and the neutron flux was obtained with the DORT code [2]. For hard-to-detect radionuclides: ^{55}Fe, ^{63}Ni, ^{59}Ni the specific and the total activity was calculated with the scaling factors method using ^{60}Co as the reference radionuclide [2]. To estimate the scaling factors, theoretical calculations, gamma spectrometric measurements and data from the literature [3, 4, 5] were used. At the decommissioning phase, the reactor block inventory is estimated based on gamma ray measurements of the waste containers and on scaling factors methods of the hard-to-detect radionuclides activities [2].

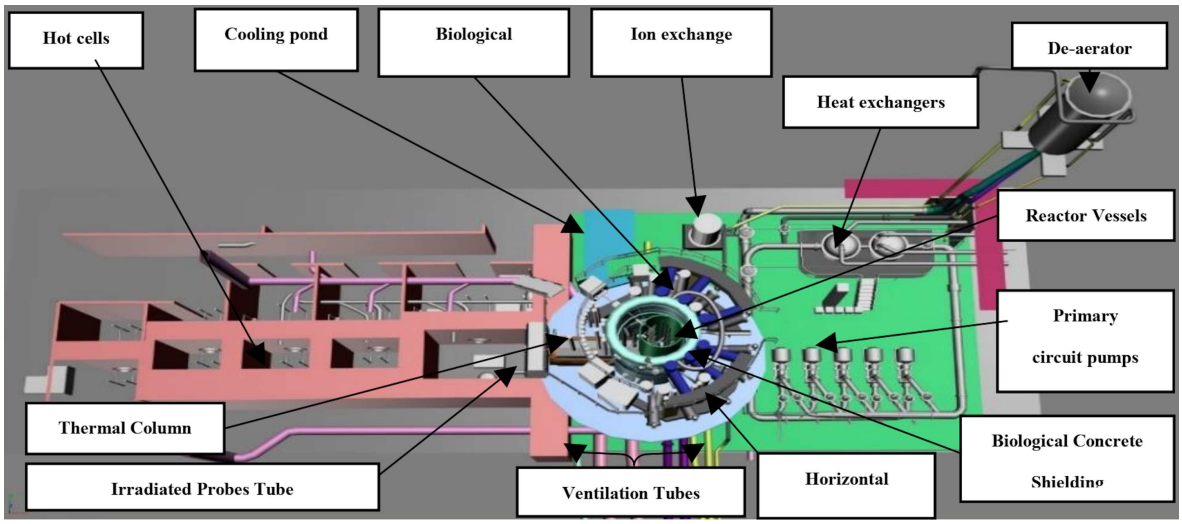

Fig. 2. Reactor overview.

3. Systems for radionuclide inventory measurement

Two main systems dedicated to the measurement of the radionuclide inventory contained in the radioactive waste packages were employed. One of them is the Ortec ISOCART system consists of: HPGe (hyper pure Germanium detector), p-type, GEM25P4 model, crystal diameter 5.74 cm, crystal length 5.22 cm; portable multichannel analyser, DigiDART type; lead collimator, 20.3 cm length, 1.6 cm thickness. The specifications guaranteed by the manufacturer are: relative efficiency 25%, resolution 1.85 keV to 1332 keV (^{60}Co) and 0.82 keV to 122 keV (^{57}Co). The software package consists of: (i) MAESTRO32 - interface between equipment and user for acquisition and analysis of spectra; (ii) ISOTOPIC32 — HPGe detector application system assuring basic and advanced procedures for accurate and consistent measurements, providing practical solution for various measurement geometries encountered among the materials from decommissioning of the nuclear facilities as it described by Gurau et al. in paper [6]. The other system is segmented gamma scanner WS-1100 — Canberra type — which allows an automatic scanning of packages with radioactive wastes. The sample is divided onto vertical

slices. Data are acquired for each of them. The transmission measurements give the estimation of average attenuation coefficient with good accuracy [7]. The non-homogeneity (in the vertical direction) of activity and density can also be assessed. The system consists in: HPGe detector, p-type, model GC4020, crystal diameter 6.1 cm, 6.4 cm length; digital analyser, model DSA1000; lead collimator, length 36.5 cm, with a rectangular slot at the front of the detector, 14 cm long and 3.6 cm high. The relative efficiency is 44.4%, 1.92 keV energy resolution at 1332 keV (^{60}Co) and 1.08 keV at 122 keV (^{57}Co) [7].

4. Results and discussion

4.1. Reactor block inventory

In the decommissioning period (2010÷2017) various aluminium components were dismantled: vertical irradiation channels, Ruther I and II irradiation loop, the channel for irradiating silicon samples, the Glantz loop protection channel and the internal reactor vessels; the channel for loading of the fuel assemblies into the reactor vessel, the lining of the cooling pond for spent fuel assemblies; the graphite thermal column (the cast iron support and wheels); the pneumatic mail from horizontal channel no. 7. The manual controller rod (2RR) made of boron carbide, stainless steel and carbon steel components (the ionisation chambers, the support sheaths for control rods, the automatic regulator bar and pipes, the de-aerator and the associated HEPA filters/pre-filters) were dismantled also. Resulted at about 78 containers of 220 l and 4 ecoRAD 50 containers with a total inventory of 9.3×10^{10} Bq (2.5 Ci). The highest activity was registered for the automatic regulator bar, 8.99×10^{10} Bq. The reactor thermal shield rings (cast iron) no. 1 and no. 2, the reactor lids and the thermal column protection (7.5 tons) were measured, decontaminated and free released. The cast iron rings no. 3÷18 (51 tons) were dismantled but not measured, therefore their contribution is not taken into account in this assessment. The contribution of concrete in the inventory was not taken into consideration as well, due to the fact that the biological protection shielding demolition only started in 2017. The graphite contribution measurements are obtained based on the gamma spectrometry analysis of samples from thermal column as well as from the thermal neutron fluxes obtained with the DORT code and theoretical calculation [2]. The ^{55}Fe, ^{63}Ni and ^{59}Ni activities were calculated using the values of the scaling factors [2]. Thus, by adding hard to detect radionuclides and graphite contribution, the total inventory of the radioactive waste (15.1 tons) resulted from reactor block components dismantling is 2.07×10^{11} Bq (5.59 Ci), as it shown in Fig. 3. The radioactive inventory will be definitely higher at the end of decommissioning due to the contribution of the cast iron rings as well as the biological concrete shielding (100 tons), thus approaching the estimated value of 4.34×10^{11} Bq (11.7 Ci) in 2012 [2]. The radionuclides weight in inventory is shown

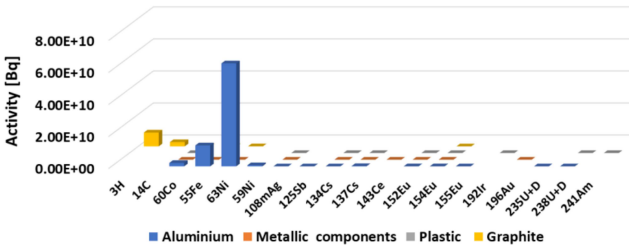

Fig. 3. Reactor block inventory - decommissioning.

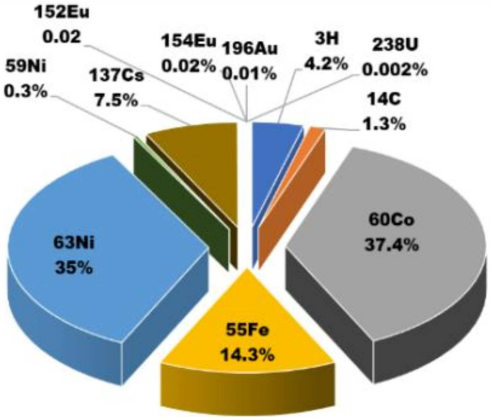

Fig. 4. Radionuclide weight in reactor block inventory.

in Fig. 4. It can be noticed that ^{60}Co (37.4%) is the dominant radioactiviy source, being present especially in the automatic regulation bar, the most active reactor component, but also in the stainless steel and aluminium components. The hard-detectable radionuclides such as ^{63}Ni (35%) and ^{55}Fe (14.3%) resulted from the activated aluminium and metallic components. ^{3}H (4.2%) and ^{14}C (1.3%) were found in the thermal column graphite and ^{196}Au (0.01%) in the ionization chambers.

The fission product ^{137}Cs is about 7.5% from the inventory and was detected in a significant amount (1.54×10^{10} Bq) in the automatic regulator bar. ^{152}Eu (0.02%) and ^{154}Eu (0.02%) were identified in the reactor internal aluminium vessels It can be noticed the presence of ^{238}U (0.002%) in the aluminium channels and sheaths of the active core. Analysis of the inventory distribution by material types (see Fig. 5) suggest, that the highest activity is concentrated in iron components (55.6%). Aluminium components contain 38.8% of the activity and

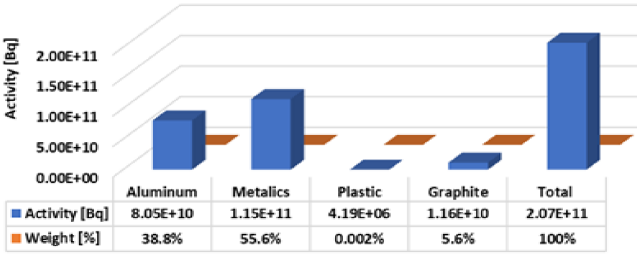

Fig. 5. Reactor block inventory on materials.

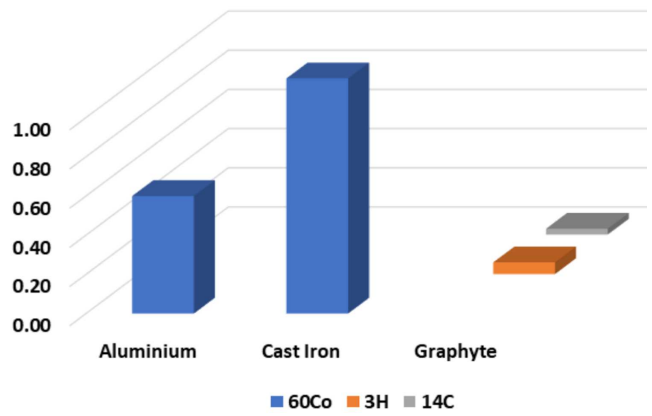

Fig. 6. Comparison between measured and estimated activity concentration in reactor block.

graphite 5.6%. Plastic share is just only 0.002%. The most important analysis regarding the reactor block inventory consists in the comparison of the aluminium and cast-iron activity concentrations, as it resulted from reactor dismantling in 2017, with those estimated in 2012 by Ionescu et. al. in paper [2]. Regarding ^{3}H and ^{14}C, the comparison was made between the activity concentrations estimated in 2012 [2] and those experimentally determined by the Enachescu et. al. in paper [8] using oxidation method in two sampling steps in oxygen atmosphere [8] for radiological characterization of the irradiated graphite from VVR-S research reactor. The results are shown in Fig. 6. Within the 5 years, the initial estimated inventory of the ^{60}Co was reduced to half the initial value. It can be notice that the ratio between measured and estimated for ^{60}Co of 0.6 in aluminium and 1.2 in cast iron shows a good agreement between the estimated and the real situation. The activity concentrations of the concrete were not compared due to the fact the biological shielding demolition began in 2017. The ratio between measured and estimated for ^{3}H of 0.06 respective ^{14}C of 0.03 in the irradiated graphite are in good agreement, taking into considering the high uncertainties in evaluation and measurement.

4.2. Hot cells inventory

Hot cells were employed mainly to the radioisotopes production for medical purposes (mainly ^{131}I, ^{99}Mo and ^{198}Au), industrial applications (^{192}Ir, sources for gammagraphs and ^{60}Co for furnaces) and for radiochemical products manufacturing. The uranium irradiation for research and silicon irradiation for electronics industry was also carried out [1]. The hot cells radioactive

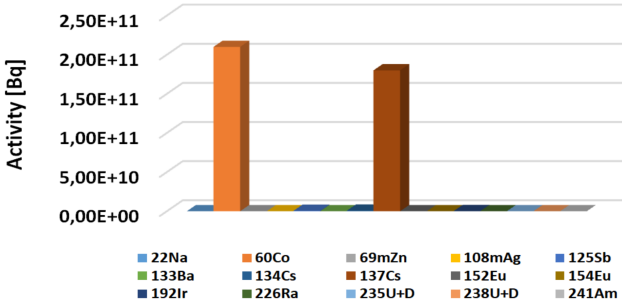

Fig. 7. Hot cells inventory - decommissioning.

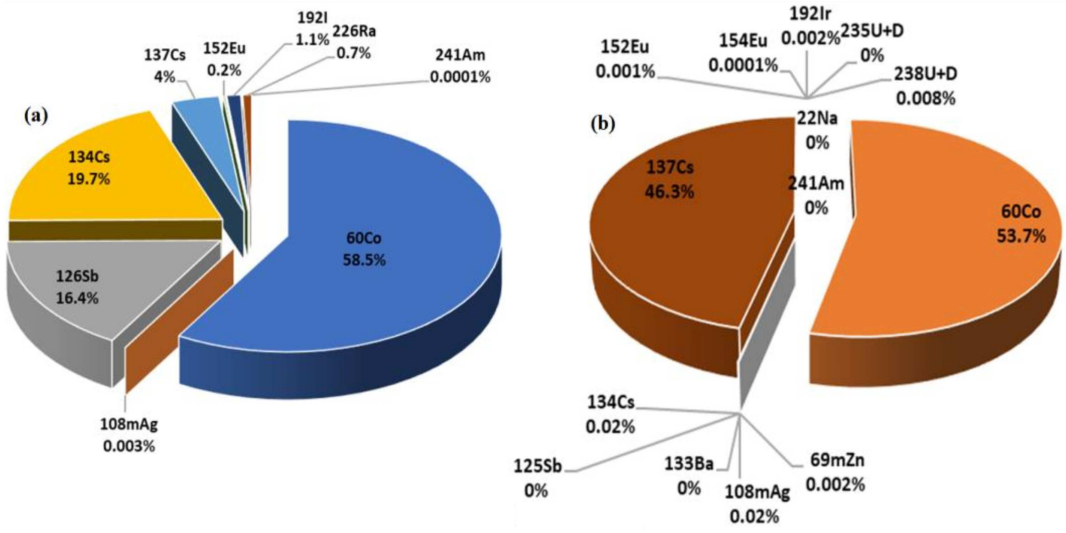

Fig. 8. Radionuclides weight in hot cells inventory (a) conservation, (b) decommissioning.

inventory, 5.5×10^{11} Bq (14.9 Ci), was estimated in 2007 before the decommissioning by Stanga et al. and presented in the VVR-S Nuclear Reactor Decommissioning, Radiological Characterisation Report [1]. The first step in hot cells decommissioning consists in the radioactive wastes and sources evacuation. The process started during the reactor conservation phase (2006÷2010) and continued in the decommissioning (2013÷2017).

The wastes (e.g. aluminium supports used to irradiate targets for radioisotopes production, plastics, rubber, vials, metallics) are put in plastic bags, labelled, measured and placed according to the dose rate value in lead containers, in containers with metallic sheets or concrete protection. The radioactive wastes inventory (see Fig. 7) is obtained based on the gamma spectrometric measurements of the containers. During 2006÷2017 were discharged from the hot cells at about 2.71 tons of radioactive waste (88 containers), 3.91×10^{11} Bq (10.57 Ci).

The radionuclide weight in the inventory is shown in Fig. 8a (conservation period) and respectively Fig. 8b (decommissioning period). Regarding the conservation phase, one can notice that the activation product ^{60}Co (58.5%) is the most abundant radionuclide, but there were also identified ^{192}Ir (1.1%) in metallic and aluminium wastes and ^{152}Eu (0.2%) in aluminium. The fission products ^{134}Cs (19.7%) and ^{137}Cs (4%) were found in all materials and ^{125}Sb (16.4%) just in aluminium. The actinides ^{226}Ra (0.7%) and ^{241}Am (0.001%) were identified in the metallic respective glass vials evacuated from the hot cells adjacent rooms. Regarding the wastes removed in decommissioning ^{60}Co (53.7%) is the most abundant. The fission products such as: ^{137}Cs (46.3%) and ^{134}Cs (0.02%) were found in all types of wastes.

^{152}Eu (0.001%) and ^{154}Eu (0.0001%) were found only in metallic wastes. Actinides have a very little percentage in the inventory and were identified in metallic wastes (^{228}U and ^{235}U) and linoleum (^{241}Am) from the adjacent room of the hot cells.

5. Conclusion

The radioactive inventory of the VVR-S nuclear research reactor is concentrated mainly in the reactor block and Hot Cells. The reactor block inventory estimated initially in 2012 based on the space distribution of the thermal neutron flux inside reactor structures and gamma ray spectrometry measurements of the samples [2] is compared with the inventory resulted from decommissioning obtained by gamma ray measurements of the waste containers. It is noticed a good agreement between the estimated and the real situation. For ^{60}Co, the key activation product, the ratio between measured and estimated is of 0.6 in aluminium and 1.2 in cast iron. For ^{3}H and ^{14}C from the graphite thermal column the ratio is 0.06 respective 0.03. The hot cells inventory as confirmed by the gamma ray measurements of the radioactive waste containers is significant and representing at about 71% of the estimated ones. The assessment of the inventory for the solid radioactive waste resulted from the decommissioning of a nuclear research reactor is essential for the establishment of the ways for radioactive waste storage and also constitutes a reference for stakeholders in the decommissioning of the similar nuclear facilities.

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