EVALUATION OF RADIONUCLIDES TRANSPORT IN CONCRETE USING SPECTROMETRIC DATA MEASURED IN THE IFIN-HH VVR-S RESEARCH REACTOR BUILDING

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Received September 26, 2018

Abstract. The VVR-S nuclear research reactor, owned by Horia Hulubei – National Institute for Physics and Nuclear Engineering (IFIN-HH), was the first research reactor using the VVR-S type soviet design it was built between 1955 and 1957, in Romania. The role of the reactor was for research and radioisotope production. VVR-S means that is a thermal neutrons reactor model S moderately cooled and reflected with distilled water, fuelled with enriched uranium. The research reactor operated until 1997. The decommissioning of the reactor has started in 2010 and is planned to be finalized in 2020. In the paper an evaluation of radionuclides transport in concrete structures from the pump room is made using measured spectrometric data.

Key words: radionuclides, transport, concrete.

1. INTRODUCTION

The VVR-S nuclear research reactor, owned by Horia Hulubei – National Institute for Physics and Nuclear Engineering (IFIN-HH), was the first research reactor using the VVR-S type soviet design in Romania. The role of the reactor was for research and radioisotope production. VVR-S means that is a thermal neutrons reactor model S moderately cooled and reflected with distilled water, fuelled with enriched uranium 10% in the beginning and 36% subsequently.

The research reactor operated until 1997 and was permanently shut-down in 2002. During his life time, it was functional for a period of 113467 h, including 2000 h at 3.0–3.5 MW power. The total power output up to 1997 was 9.59 GWd. It had a utilization factor of 65 % (approximate 9510 effective days of operation)

with an average of 1 MW thermal power. It sustained a thermal neutron flux of max. $2x10^{13}$ n/cm^{2}sec.

The first criticality was attained on July 29, 1957 with a fuel loading of 4.5 kg of 235U. Its last shut down was on July 1997. All of the fuel (10% 235U until 1984, and 36% 235U after this date) was removed from reactor core to the Cooling Pond (Reactor Hall) and the basins of Spent Nuclear Fuel Storage (Building No. 20).

The decommissioning of the reactor has started in 2010 and is planned to be finalized in 2020, being the first nuclear research reactor that has started the decommissioning project from the South-East of Europe [1].

Radioisotope production generated a significant contamination in the reactor main building, ventilation system and radioactive leakage drainage, overflow and collecting system. Major radioactive contaminants generated by this activity, with the half life higher than one year, are: Co-60, Cs-134, Cs-137, Sr-90, Eu-152, U-238 and Am-241 [1].

2. RADIOLOGICAL CHARACTERIZATION OF ROOM 30 AND 31

2.1 Sampling of radionuclides

The main activation products found when the reactor was shut down:

- ^{55}Fe, ^{60}Co, ^{59}Ni, ^{63}Ni, ^{39}Ar, ^{94}Nb in cast iron and iron,
- ^{3}H, ^{14}C, ^{41}Ca, ^{55}Fe, ^{60}Co, ^{152}Eu, ^{154}Eu in concrete,
- ^{3}H, ^{14}C, ^{152}Eu, ^{154}Eu, in graphite. [2]

Regarding the iron and cast iron, 55Fe and 60Co are the most important part of the radioactive inventory for the first ten years after shut down. Most of these isotopes are disintegrated during the next fifteen years. The main activation component is given by 59Ni, 63Ni and 94Nb, which have greater half-life. Regarding the concrete and graphite, the most important contribution for long term is given by 14C, 41Ca and the europium isotopes.

The specific types and numbers of measurements are specified for every area listed below. The alpha, beta and gamma activity is measured for all the samples (in a fixed geometry) in order to establish if an analysis is necessary to identify radionuclides and to determine the specific activity. The radionuclides will be identified only for those samples for which the activity exceeds the minimum detectable activity –MDA-specified.

The large samples can be divided into sub-samples. The concentration of each radionuclide is determined only for one or two sub-samples. For the other sub-samples only the gross activity is determined. The ratio of the nuclide's

specific activity concentrations to the gross activity can be used to infer the activity concentrations for all the other sub samples.

Where alpha contamination is found, the laboratory will perform an alpha spectroscopy analysis to determine the alpha emitting radionuclides. Where beta activity is observed, the gamma spectroscopy results will be compared to the results of the gross beta analysis.

Where beta activity detected is not associated with gamma activity, samples will be analyzed to determine the activity concentration of the following beta emitting radionuclides: 3H, 14C, 36Cl, 99Tc, 63Ni, 55Fe, 90Sr, 90Y.

The size of samples (volume, mass, geometry) must be selected in such a way as to give results of laboratory measurements with a minimum 95% confidence level.

Two floods occured in the pumps room in 1972 due to an overflow from the 300 m3 tank into the buffer reservoir and the pump room. The influence of these floods is pointed out by the results of the radiological characterization for rooms 31 and 30

Room 30 is fully and highly contaminated. Contamination penetrated into the wall and the depth penetration is about 2 mm.

- Nuclide vector: Cs-134, Cs-137 and Co-60. Cs-134 is the predominant radionuclide.

- Contamination level: max. 103.3 Bq/cm2.

This room must be decontaminated taking into account the contamination depth penetration.

Room 31 is highly contaminated. Contamination penetrated into the floor and the depth penetration is about 2 cm. Contamination is concentrated behind pumps and their pedestal.

-Nuclide vector: Co-60 and Cs-137. Co-60 is the predominant nuclide.

-Contamination level: max. 23.8 Bq/cm2.

-Bulk contamination: max. 20.7 Bq/g.

This room must be decontaminated taking into account the contamination depth penetration.

2.3 Operation procedures chosen for measurements

Room below the Reactor (room 30) (Fig. 1):

- Scan the floor and walls.
- Record the average values.
- Identify the hot spots; take measurements of the dose rate with LB 6006 and in situ measurements.
- Survey alpha, beta, gamma gross activity for contaminated areas.

- Smear a 100 cm^2 surface where contamination is found (alpha, beta, gamma gross activity).
- Take two smears at random from the floor and walls to determine alpha, beta, gamma gross activity. If hot spots are found, take samples from these areas.
- Scan contaminated areas to determinate alpha activity. If alpha activity is found, take in situ measurements.
- If $\beta\gamma$ activity is found, submit one smear sample for γ spec. If alpha contamination is found, submit one smear sample for α spec.
- Scan and smear the ceiling (made of cast iron sustaining plate) and take 4 smears at random.
- Drill the plate and submit some samples from the location with the highest activity for γ spectrometry.
- Scan the pipes and take smears from the flanges area.
- Measure lead in paint with XRF.

Pumps Room (room 31) (Fig. 2):

- Scan and smear the floor and walls of the access corridor (up to the 2 m above the floor).
- Scan and smear the room's floor.
- Scan and smear the room's equipment.
- Record the areas with the highest dose rate.
- Scan the walls and ceiling.
- Take a smear from each wall and the area with the highest activity.
- Open the pipes flanges from the five water pumps of the primary circuit.
- Take smears and samples from waste if found.
- Prepare a complex sample and analyze it by γ spectrometry.
- Take a sample from the ion exchange resin. Analyze it to detect heavy metals by γ spectrometry.
- Take an oil sample from the gearing of the pumps. Analyze it to detect PCB, heavy metals by γ spectrometry.
- Check the flanges' clamps and walls anchors of the pipes for asbestos.
- Take a sample from the stainless steel drain sewer. Analyze it.
- Drill a 50 mm diameter core with a depth of 100 mm from the most contaminated fixed area from the pumps concrete bedplate.
- Drill a 50 mm diameter core with a depth of 100 mm from the most contaminated fixed area from the concrete of the drain sewer.
- Drill a 50 mm diameter core with a depth of 100 mm from the most contaminated fixed area from the pumps room's floor.
- Scan and smear the cores. Mark the location with the highest dose rate.

- Take paint samples from walls, ceiling and equipment,
- Measure lead in paint with XRF.

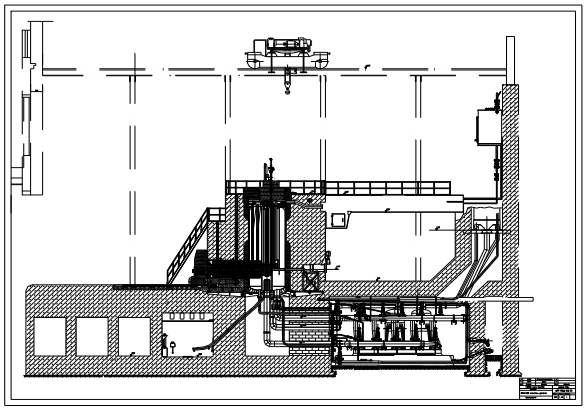

Fig. 1 Room 30 position in Nuclear Reactor – profile view [2]

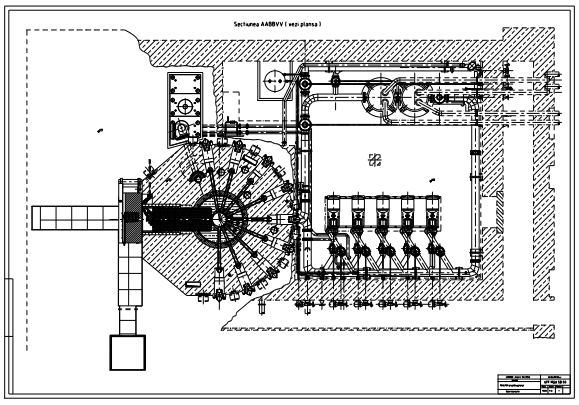

Fig. 2 Room 31 position in Nuclear Reactor – above view [2]

3. METHODOLOGY OF MEASUREMENTS

3.1 Data Conversion

Radiological survey data is usually obtained in units, such as counts per unit time, which have no intrinsic meaning relative to the guideline values. Therefore, the survey data from field and laboratory measurements are converted to units, which will enable comparisons. Standard units used for expressing survey findings are:

- Surface Contamination - Bq/cm^2 (disintegrations per second per cm^2)
- Radionuclide Concentration - Bq/g (disintegrations per second per gram)
- Dose Rat - μSv/h (microsievert per hour)
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The methodolgy of measurements is presente in following diagram. (Fig.3).

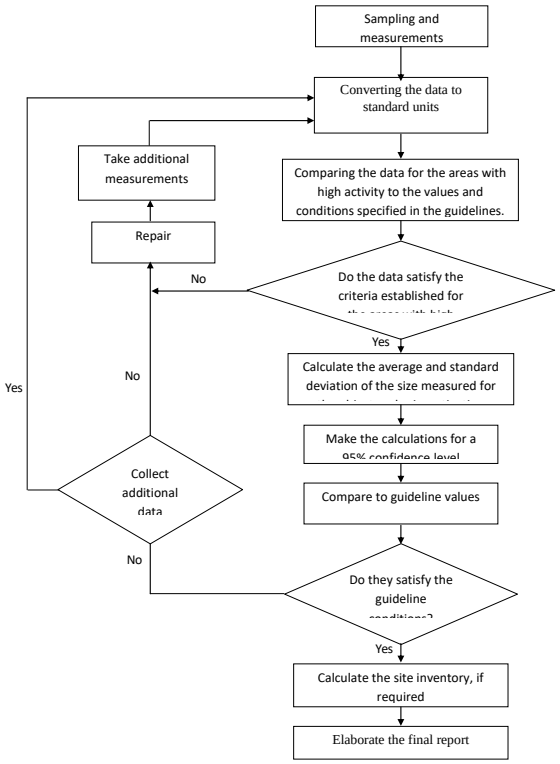

Fig. 3 Diagram of the process for interpreting and comparing the monitoring data to the release levels [3]

3.2 Surface Activity

A measurement for surface activity is performed over an area, represented by the sensitive surface area of the detector. If the instrument display is in count rate, i.e. counts per second, the conversion to Bq/cm^2 is performed by:

$$A_s = (n - n_B) / (E \bullet W) \quad [4] \quad (1)$$
$$[Bq/cm^2] = [counts/second]/ ([counts/decay] \bullet [cm^2])$$

where

A_s = surface activity, in Bq/cm^2;
N = count rate, in counts per second;
n_B = background count rate, in counts per second;
E = efficiency, in counts per disintegration (counts/decay);
W = surface of the detector window, in cm^2.

For a technique using an integrated count on a digital instrument the conversion is:

$$A_s = (N-N_B) / (t \bullet E \bullet W) \quad (2)$$
$$[Bq/cm^2] = [counts] / ([seconds] \bullet [counts/decay] \bullet [cm^2])$$

where

N = total counts recorded by the measurement;
N_B = count during recording period, due only to background levels of radiation;
t = time period over which the count was recorded, in seconds.

The level of removable activity collected by a smear is calculated in the same manner:

$$A_s = (N-N_B) / (t \bullet E \bullet S \bullet F) \quad (3)$$
$$[Bq/cm^2] = [counts] / ([seconds] \bullet [counts/decay] \bullet [cm^2])$$

where

S = the area smeared, in cm^2;
F = removal factor (unitless).

3.3 Radionuclide Concentration

To determine the radionuclide concentration in Bq/g, the calculation performed is:

$$A = (N\text{-}N_B) / (t \bullet E \bullet M) \quad [5] \quad (4)$$

[Bq/g] = [counts] / ([seconds] • [counts/decay] • [g])

If the analytical procedure includes a wet chemistry separation, it will also be necessary to correct for the fractional recovery (R), determined by a spike or tracer added to the sample.

$$A = (N\text{-}N_B) / (t \bullet E \bullet M \bullet R) \quad (5)$$

[Bq/g] = [counts] / ([seconds] • [counts/decay] • [g])

where

M = weight of sample, in grams

3.4 Dose Rate

If an instrument, such as a pressurized ionization chamber or a dose rate meter is used for measuring exposure rate, the instrument reading will be directly in the desired exposure rate units (μSv/h). A gamma scintillation or GM detector with a count rate or digital scaling instrument provides data in units of counts per second or per some preset time, respectively. Conversion to μSv/h is accomplished, using calibration factors developed for the specific instrument and survey site. The background exposure rate is then subtracted from their total to determine the net level. This net level is compared with the guideline value.

$$D = (n – n_B) \bullet C \quad [6] \quad (6)$$

[μSv/h] = [counts/second] • [[μSv/h]/[counts/second]]

or

$$D = (N – N_B) \bullet C / t$$

[μSv/h] = [counts] • [[μSv/h]/[counts/second]] / [seconds]

where

D = dose rate, in micro sievert per hour;

C = calibration factor.

4. DATA MEASUREMENTS RESULTS

We determined the depth penetration of the contamination. Thus, we extracted concrete cylinders (diameter=5cm) from the floor in room 31. Samples were cut from cylinders and then measured by gamma spectrometry [7]. The the depth penetration is about 2 cm. Measurement results are given in Table 1.

In room 30, layers were removed from the wall. After each layer removal, we measured the surface activity. Results are given at the end of Table 2. In this case, the depth penetration is about 2 mm.

Table 1.
Contamination depth penetration-Room 31

Point no. (see App.17)	Sample code	Depth (cm)	Specific activity (Bq/g)		
			Co-60	Cs-137	Cs-134
CDP1	1	0.00	0.00	0.00	0.00
CDP2	2	0.00	0.00	0.00	0.00
CDP3	3	0.00	13.10	1.69	0.00
CDP4	4	0.00	0.18	0.05	0.00
CDP5	5	0.00	20.68	4.39	0.40
CDP6	6	0.00	11.25	2.48	0.00
CDP7	7	0.00	0.00	0.00	0.00
CDP8	8	0.00	0.00	0.00	0.00
CDP9	9	0.00	0.00	0.00	0.00
CDP10	10	0.00	0.00	0.00	0.00
CDP11	11	0.00	0.00	0.00	0.00
CDP12	12	0.00	0.00	0.00	0.00
CDP5	136	10.0	0.00	0.00	0.00
CDP5	388	2.00	0.00	0.00	0.00
CDP5	389	4.00	0.00	0.00	0.00
CDP5	390	7.00	0.00	0.00	0.00

Table 2
Contamination depth penetration –Wall of the Room 30

Radiation Monitor E-600 SN02996–Probe HP-210T SN727425				
Depth (mm)	Background (cps)	Counting rate (cps)	Contamination (Bq/cm^2)	Remarks
0	2.7	22.4	6.57	Point W9 (Middle of cell-9, High=1.5m)
1	2.4	6.0	1.20	Layer of 1 mm was removed from the wall.
2	2.3	3.5	0.4	Another layer of 1 mm was removed from the wall.

Remarks:

- Dose-rate measurements were performed at 1 m above the floor.
- Measurement points from walls correspond to the middle of the cell at 2±0.5 m hight.

From Table 3, we identified 10 areas of elevated activity (surface activity higher than exclusion level) in the Room 30.

Table 3.
Areas of elevated activity in the Room 30

Point no.	Surface (cm2)	Smear code	Nuclide vector	Surface activity (direct measurements) (Bq/cm2)	Surface activity (smears) (Bq/cm2)	Removal factor
F6	~100	-	Cs-134, Co-60,Cs-137	4.71±0.9	-	-
F7	~100	-	Cs-134, Co-60,Cs-137	3.52±0.7	-	-
F8-1	~100	349	Cs-134, Co-60,Cs-137	11.90±2	0.99±0.2	0.08
F9-1	~75	-	Cs-134, Co-60,Cs-137	41.20±8	-	-
F9-2	~75	-	Cs-134, Co-60,Cs-137	80.00±16	-	-
F9-3	~75	463	Cs-134-85%, Co-60-15%	103.33±20	12.25±2.4	0.12
F10	~100	350	Cs-134, Co-60,Cs-137	6.10±1.2	1.42±0.28	0.23
F11	~100	-	Cs-134, Co-60,Cs-137	10.66±2	-	-
F12	~150	351	Cs-134-79%, Co-60-18%, Cs-137-3%	22.09±4	12.48±2.4	0.56
W9	~50	459	Cs-134, Co-60,Cs-137	6.57±1.3	0.1±0.02	0.02

In the Room 31, we identified 5 areas of elevated activity (surface activity higher than exclusion level). These areas are characterized in Table 4

Table 4.
Areas of elevated activity in the Room 31

Point no.	Surface (cm^2)	Smear code	Sample code	Specific activity (Bq/g)		Surface activity (direct measurements) (Bq/cm2)	Surface activity (smears) (Bq/cm^2)	Removal factor
				Co-60	Cs-137			
F5.1-1	~200	362	-	-	-	4.17±0.9	0.96±0.2	0.23
F4.1-2	~200	363	-	-	-	23.77±4.6	0.55±0.1	0.02
CDP 3	-	-	3	13.10±2.6	1.69±0.4	-	-	-
CDP 5	-	-	5	20.68±4.2	4.39±0.8	-	-	-
CDP 6	-	-	6	11.25±2.2	2.48±0.5	-	-	-

In Fig. 4-9, spectographic analysis are presented related to the 6 probes taken during drills in the concrete floor of room 31.

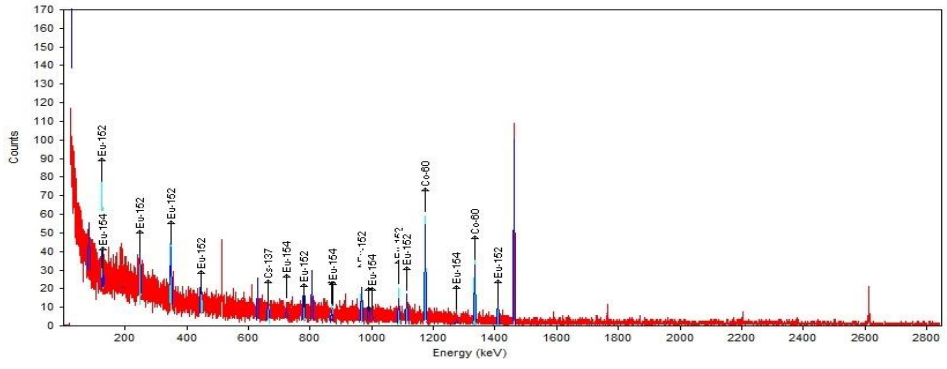

Figure 4 – Probe 1 spectrographic analysis from concrete floor drilling in room 31

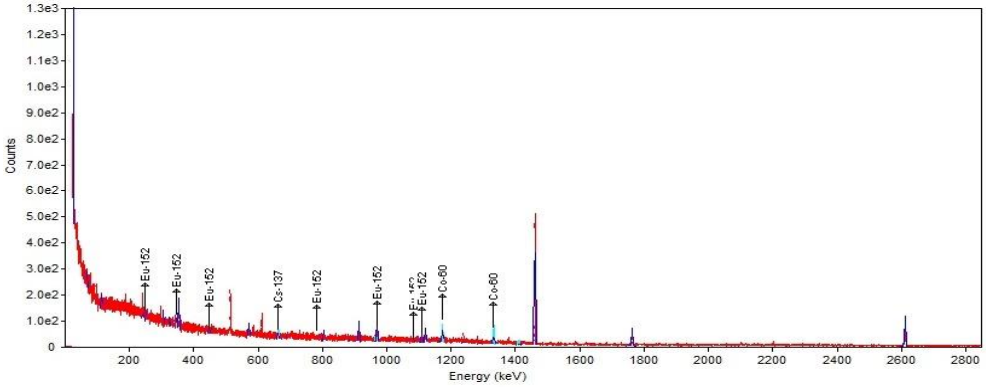

Figure 5 – Probe 2 spectrographic analysis from concrete floor drilling in room 31

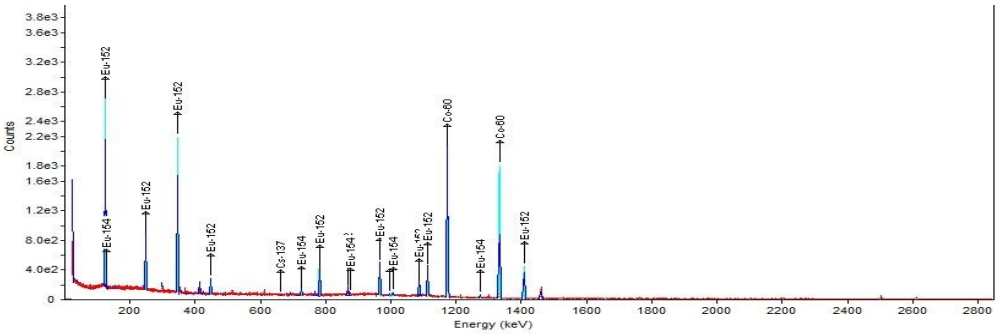

Figure 6 – Probe 3 spectrographic analysis from concrete floor drilling in room 31

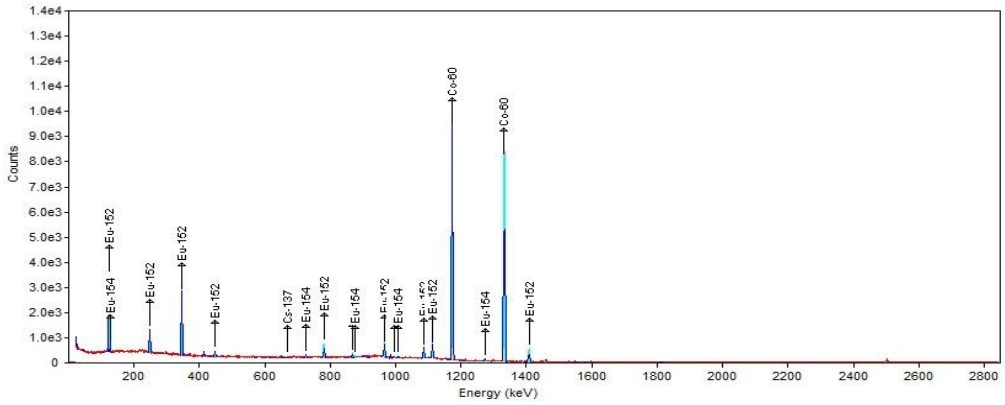

Figure 7 – Probe 4 spectrographic analysis from concrete floor drilling in room 31

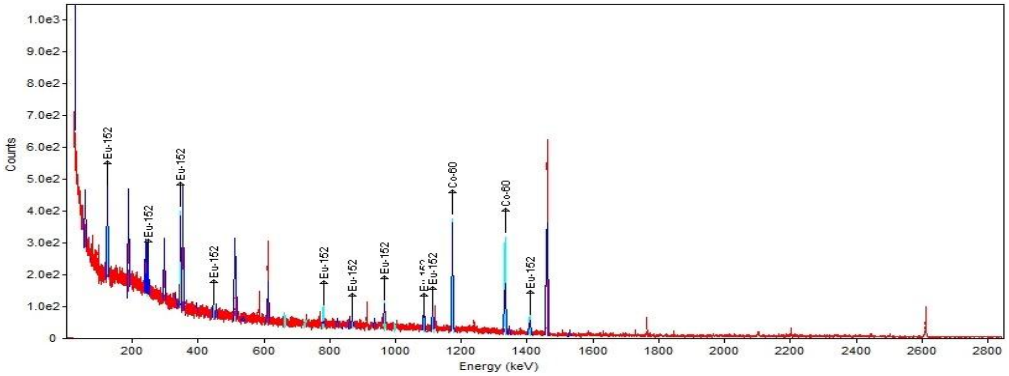

Figure 8 – Probe 5 spectrographic analysis from concrete floor drilling in room 31

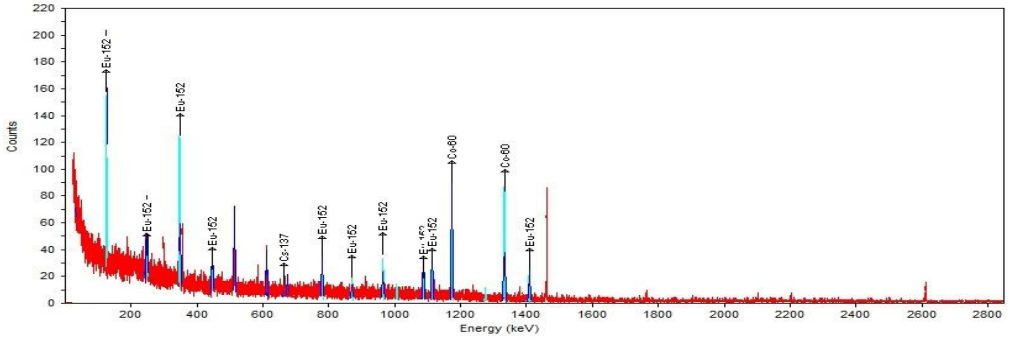

Figure 9 – Probe 6 spectrographic analysis from concrete floor drilling in room 31

5 CONCLUSION

The data input used in evaluation cosisted of the following:

- Potential radioactive contaminants: beta-gamma emitters (especially Co-60, Cs-134, Cs-137);
- Guideline value for beta-gamma emitter:
  - Surface activity: 3 Bq/cm^2 (Co-60, Cs-137, Cs-134)
  - Specific activity: 1.0 Bq/g-Co-60; 0.8 Bq/g-Cs-137; 0.5 Bq/g-Cs-134.

Radionuclide transport resulte in the followin situation:

- Room 30 is fully contaminated with Cs-134, Co-60 and Cs-137. We identified 10 areas of elevated activity in this room.
- Room 31 is highly contaminated with Co-60 and Cs-137. Contamination is concentrated within area located behind pumps (cells 1.1, 2.1, 3.1, 4.1, 5.1). In this area, we have bulk contamination due to the migration of nuclides into concrete.

As a result of this analysis it can be concluded that the rooms 30 and 31 are highly contaminated and therefore they must be decontaminated.

This task can be acomplished as follows:

- In room 31 a concrete layer of about 2 cm must be removed from the floor.
- In romm 30, a concrete layer of about 2 mm must be removed from the walls.

Acknowledgements

The work from this paper was performed under an AOSR Research Grant.
The authors would like to express their highest appreciation for the great support received from all the distinguished members and all the professional staff of the Academy of Romanian Scientists.

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