DOSE RATE AND RISKS ASSESSMENT FOR VVR-S RESEARCH REACTOR DEPLETED URANIUM WORKSHOP

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Abstract. The IFIN-HH VVR-S type Nuclear Research Reactor is currently in the decommissioning phase. During the operation period inside of the reactor building a workshop for mechanical processing of depleted uranium blocks was used. The purpose was containers manufacturing for biological shielding of Cs-137 and Co-60 sources. The dose rate and associated risk for workers performing clean-up and decontamination of the workshop was assessed using RESRAD Build 3.5 code. As a result of the workshop radiological characterization performed in 2007 it was revealed the presence of U-238 as dust particles embedded in the oil grease. The main risk for workers contamination was internal by aerosols inhalation as well as external by hand touching of the contaminated surfaces. The time for clean-up and decontamination operation was 2 weeks, and therefore the estimation was made for this period.

Keywords: dose rate evaluation, risk assessment, clean-up, decommissioning, depleted uranium.

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1. Introduction

The VVR-S nuclear research reactor was used in IFIN-HH for research in physics, biophysics, and radiochemistry and for radioisotope production. It was operated at a nominal thermal power of 2 MW, maximum neutron flux of $2x10^{13}$ n/cm²s; It produced 9.59 GWd thermal energy using distilled water as coolant, moderator, and reflector. The Reactor used two types of fuel: EK-10, (low-enriched fuel, uranium dioxide enriched by 10% U-235 isotope, dispersed in magnesium oxide matrix) and S-36, high-enriched fuel, uranium dioxide enriched 36% by U-235 isotope, dispersed in aluminium matrix. The main purpose of reactor was radioisotope production for medicine and industrial and applications such as I-131, Mo-99 and Au-198 for medical application or Ir-92 sources for gamma-graphs and Co-60 for furnaces The VVR-S nuclear research reactor was shut down in 1997, after 40 years of operation and it is now in decommissioning phase

2. Uranium health effects

According to IAEA provisions [1], uranium is a naturally occurring radioactive element, which in the pure form it is a silver-coloured heavy metal, the same as lead, cadmium, and tungsten. It is very dense, about 19 grams per cubic centimetre, 70% more dense than lead. It is so dense a small 10-centimetre cube would weigh

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20 kilograms. In its natural state, it consists of three isotopes (U-234, U-235 and U-238). Other isotopes that cannot be found in natural uranium are U-232, U-233, U-236 and U-237. The Table 1 shows the fraction by weight of the three isotopes in any quantity of natural uranium, their half-lives, and specific activity. The half-life of a radioactive isotope is the time taken for it to decay to half of its original amount of radioactivity. Uranium is a Low Specific Activity material. The specific activity is activity per unit mass of a radionuclide used as a measure of how radioactive a radionuclide material [1].

Table 1) Uranium isotopes [1]

Isotope	Relative Abundance by Weight	Half Life (Years)	Specific Activity
			[Bq/mg]
U-238	99.28%	451000000	12.4
U-235	0.72%	710000000	80
U-234	0.0057%	247000	231000

In nature, uranium isotopes are typically found in radioactive equilibrium (i.e. the activity of each of the radioactive progeny is equal to the activity of the uranium parent isotope) with their radioactive decay products. Decay products of U-238 include thorium-234 (Th-234), protactinium-234 (Pa-234), U-234, Th-230, radium-226 (Ra-226), radon-222 (Rn-222), polonium-218 (Po-218), lead-214 (Pb-214), bismuth-214 (Bi-214), Po-214 Pb-210 and Po-210. Decay products of U-235 include Th-231, Pa-231, actinium-227 (Ac-227), Th-227, Ra-223, Rn-219, Po-215, Pb-211, Bi-211 and thallium-207 (Tl-207). Isotopes of natural uranium decay by emitting mainly alpha particles. The emission of beta particles and gamma radiations are low. The Table 2 shows the average energies per transformation emitted by U-238, U-235 and U-234. [1]

Table 2)	Uranium	disintegration	(decaying)	types [1]
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Isotope	Average Energy Emitted Per Transformation		
	[MeV/Bq]		
	Alpha	Beta	Gamma
U-238	4.26	0.01	0.001
U-235	4.47	0.048	0.154
U-234	4.84	0.0013	0.002

Uranium is used primarily in nuclear power plants; most reactors require uranium in which the U-235 content is enriched from 0.72% to about 3%. The uranium remaining after removal of the enriched fraction is referred to as depleted uranium. It contains about 99.8% U-238, 0.2% U-235 and 0.0006% U-234 by mass. For the same mass, depleted uranium has about 60% of the radioactivity of uranium [1]. Depleted uranium may also result from the reprocessing of spent nuclear reactor

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fuel. Under these conditions another uranium isotope, U-236 may be present together with exceedingly small amounts of the transuranic elements: plutonium, americium and neptunium and the fission product technetium-99. The increase in the radiation dose from the trace amounts of these additional elements is less than 1%. This is insignificant with respect to both chemical and radiological toxicity. Depleted uranium has several peaceful applications: counterweights or ballast in aircraft, radiation shields in medical equipment used for radiation therapy and containers for the transport of radioactive materials.

Individuals can be exposed to depleted uranium in the same way they are routinely exposed to natural ones, i.e. by inhalation, ingestion, and dermal contact (including injury by embedded fragments). Inhalation is the most likely route of intake during or following the use of depleted uranium when it is resuspended in the atmosphere by wind or other forms of disturbance. Accidental inhalation may also occur because of a fire in a depleted uranium storage facility.

Ingestion could occur in large sections of the population if their drinking water or food became contaminated with depleted uranium. In addition, the ingestion of soil by children is also considered a potentially important pathway.

Dermal contact is considered a relatively unimportant type of exposure since little of the depleted uranium will pass across the skin into the blood. However, depleted uranium could enter the systemic circulation through open wounds or from embedded depleted uranium fragments.

Regarding body retention, most (>95%) uranium entering the body is not absorbed but is eliminated via the intestinal transit. From blood absorbed uranium, approximately 67% will be filtered by the kidney and excreted in the urine in 24 hours. Typically, between 0.2 - 2% of the uranium in food and water is absorbed by the gastrointestinal tract [2]. Soluble uranium compounds are more readily absorbed than those which are insoluble. Potentially depleted uranium has both chemical and radiological toxicity with the two important target organs being the kidneys and the lungs. Health consequences are determined by its physical and chemical nature and exposure duration. Long-term studies of workers exposed to uranium have reported some impairment of kidney function depending on the level of exposure. However, there is also some evidence that this impairment may be transient, and that kidney function returns to normal once the source of excessive uranium exposure has been removed. Insoluble inhaled uranium particles $(1-10 \ \mu m)$, tend to be retained in the lung and may lead to irradiation damage and even lung cancer if a high enough radiation dose results over a prolonged period. Direct contact of depleted uranium metal with the skin for several weeks, is unlikely to produce radiation-induced erythematic (superficial inflammation of the skin) or other short-term effects. Follow-up studies of veterans with embedded fragments in the tissue have shown

detectable levels of depleted uranium in the urine, but without apparent health consequences.

Another important aspect concerning uranium is chemical toxicity and radiological dose. The public ingestion of soluble uranium compounds should not exceed the tolerable intake of 0.5 μ g/kg of body weight per day. Insoluble uranium compounds are significantly less toxic to the kidneys, and a tolerable intake of 5 μ g/kg of body weight per day is applicable. Inhalation of soluble or insoluble depleted uranium compounds by the public should not exceed 1 μ g/m³ in the respirable fraction [2]. The limit is derived from renal toxicity for soluble uranium compounds, and from radiation exposure for insoluble uranium compounds. Excessive worker exposure to depleted uranium via ingestion is unlikely in workplaces where occupational health measures are in place. Occupational exposure to soluble and insoluble uranium compounds for 8-hour time weighted average should not exceed 0.05 mg/m³ [2]. This limit is also based both on chemical effects and radiation exposure.

3. Workshop clean-up activities

The manufacturing workshop of Cs-137 and Co-60 sources biological shielding was located in the Reactor Building (rooms 43-47) and used between 1980-1995. Initially those rooms hosted a radiochemistry laboratory fitted with ventilated enclosures connected to the technological ventilation and special drainage system for radioactive liquids.

The shielding was manufactured of depleted uranium bocks imported from UK. The depleted uranium blocks processing was made with a lathe machine using a flow of oil for protection against self-ignition (see Fig. 1). To prevent the workers irradiation, the workshop has been fitted with a supplementary HEPA ventilation system (see Fig. 2).



Fig. 1. The contaminated lathe machine



Fig. 2. HEPA ventilation system

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During the process, the aerosols as well as heating gases were produced and consequently the workers potential irradiation was caused by their release as well as liquid drops resulted from cutting. The results of the workshop radiological characterization performed in 2007, confirmed the presence of U-238 small traces embedded in the oil grease. The radiological risks could arise from the inhalation of U-238 airborne aerosols as well as from external contamination caused by skin contact with oil films containing uranium particles.

In 2012, within the Reactor decommissioning, rooms 43 - 47 were cleaned-up. Prior to decontamination, the surfaces were dusted using an industrial vacuum machine (see Fig 3). The workers could have been internally exposed by contaminated dust inhalation.



Fig. 3. Industrial vacuum machine fitted with HEPA filtration

Then, the surfaces decontamination was performed using the DeconGelTM decontaminant, a polymer used in the nuclear field to confine the radioactivity. The gel was spread on the surfaces for 24 hours and then removed (see Fig. 4(a) and Fig. 4(b)). The workers contamination could be from caused by skin contact with oil films containing uranium particles. The gel was measured, was put into containers along other dismantling scraps and transported to the IFIN-HH Department of Radioactive Wastes Management for intermediate disposal (see Fig. 5).



Fig. 4. a. Floor decontamination



Fig 4. b. Wall decontamination



Fig.5. Containers with very low-level radioactive wastes

4. Modelling Dose Rate/Risk in Room 43 Clean-up Operations

4.1. Radiological conditions prior to clean-up

Prior to decontamination a thorough characterization of the room 43 (see Fig. 6) was performed. For this purpose, it was taking into account the object inventory (see table 3). A systematic gamma scan was performed using a Monitor LB 123 – Probe LB 1231 No. 6008 to determine all the significant hotspots location (higher than 1 Bq/cm², coloured in red) and their corresponding surface activity (see Fig. 7). The results are presented in Table 4 [3]. Additionally, in the hot spots dose rates measurements were performed using Monitor LB 123 – Probe LB1236 No. 180851-2761 (See Table 4). The surface contamination was completely removed after decontamination by applying the DeconGel only once due to low initial levels.



Fig. 6. Images from the processing room 43

Table 3) Object inventory in room 43

No.	Denomination	Quantity
1	Suspended rack	1
2	Box	1
3	Metallic cylinder	1
4	Suspended rack	1
5	Vats	2
6	Plastic bag (with wooden wastes)	1



Fig. 7. Measuring point in processing room 43 [3]

Measuring	Surface	Smear code	Surface	Removing	Dose rate
point	Activity		Activity	Factor	$[\mu Sv/h]$
-	$[Bq/cm^2]$		$[Bq/cm^2]$		
43-1	0.58	409	0.006	0.01	0.14
43-2	2.41	410	0.005	0.00	
43-3	0.69	411	0.003	0.00	
43-4	0.36	412	0.002	0.01	
43-5	6.90	413	0.008	0.00	
43-6	1.02	414	0.003	0.00	
43-7	7.60	415	0.027	0.00	
43-8	2.10	416	0.015	0.01	
43-9	2.80	417	0.011	0.00	
43-10	1.73	418	0.007	0.00	
43-11	0.60	419	0.002	0.00	
43-12	0.50	420	0.000	0.00	0.11
43-13	2.40	421	0.002	0.00	
43-14	0.54	422	0.003	0.01	
43-15	1.30	423	0.006	0.00	
43-16	9.40	424	0.007	0.00	0.12
43-17	9.10	425	0.009	0.00	
43-18	0.70				
43-19	0.32				
43-20	0.24				0.07
43-21	0.80				
43-22	1.00				0.13
43-23	0.76				
Average	2.3413				
Activity					

Table 4) Contamination measurements on the floor of processing room 43

4.2. Risk assessment methodology

The dose rate and associated risk for the workers performing clean-up and decontamination of the workshop was assessed using RESRAD Build 3.5 code. RESRAD is a computer model designed to estimate radiation doses and risks from RESidual RADioactive materials (see Fig. 8) applied to over 300 sites in the U.S. and other countries.



Fig. 8 RESRAD computer code package [4]

RESRAD features (see Fig. 9) [5] considers: the external exposure, inhalation of dust and radon and ingestion of soil/dust. Up to 10 sources and 10 receptors can be modelled. Sources geometry can be point, line, plane, or volume. Building can be any structure composed of up to three compartments. Radioactive contamination can be on surface or in building material. Exposure scenarios considered include building occupancy (residential use and office worker) and building remediation (decontamination worker and building renovation worker).



Fig. 9. Exposure Pathways Incorporated into the RESRAD-BUILD Computer Code [5]

Seven exposure pathways are considered in the RESRAD-BUILD code e.g.: the external exposure directly from the source; external exposure to materials deposited on the floor; external exposure due to air submersion. Inhalation of airborne radioactive particulates and aerosol indoor radon progeny and tritiated water vapor are also considered. It is taking into account the inadvertent ingestion of radioactive material directly from the source and ingestion of materials deposited on the surfaces of the building compartments. The external radiation penetrating the walls, ceilings, or floors is calculated based on the input for shielding material type, thickness, and density. A shielding material can be specified between each sourcereceptor pair, by user (from eight different material types). Internal (inhalation and ingestion) exposures are calculated based on an air quality model that considers the air exchange between rooms and with outdoor air. Two direct exposure models based on the geometrical type of sources are used. The model used for the point and line contamination is a simple dose integral method. Model for area and volume sources are based on a semi-infinite slab source, with corrections for geometrical factors. The volume source can have up to five layers, with any one layer being contaminated. The area source is treated as a volume source of small thickness (0.01 cm) with unit density.

4.3. Calculation of the intake rates and time integrated cancer risks

RESRAD can calculate lifetime cancer risks resulting from radiation exposure. The radiation risk can be computed by using the U.S. Environmental Protection Agency (EPA) risk coefficients with the exposure rate (for the external radiation pathways) or the total intake amount (for internal exposure pathways).

The EPA risk coefficients are estimates of risk per unit of exposure to radiation or intake of radionuclides that use age- and sex-specific coefficients for individual organs, along with organ-specific dose conversion factors (DCF). The EPA risk coefficients are characterized as best-estimate values of the age-averaged lifetime excess cancer incidence risk or cancer fatality risk per unit of intake or exposure for the radionuclide of concern. Detailed information on the derivation of EPA risk coefficients and their application can be found in several EPA documents (EPA 1991a, b; 1994, 1997; Eckerman et al. 1999) [5]. The methodology used in the RESRAD code for estimating cancer risks follows the EPA risk assessment guidance (EPA 1997) and is presented in the following section.

Intake rates calculated by the RESRAD code are listed in the INTRISK.REP report generated after each execution. They are listed by radionuclide and pathway and correspond to specific times. Intake rates for inhalation and ingestion pathways are calculated first for all the principal radionuclides and then multiplied by the risk coefficients to estimate cancer risks.

For inhalation and soil ingestion pathways (p = 2 and 8, respectively), the intake rates (Bq/yr or pCi/yr) can be calculated by using the following Equation (1) [4]:

$$(Intake)_{j,p}(t) = \sum_{i=1}^{M} ETF_{j,p}(t) \times SF_{i,j}(t) \times S_i(O) \times BRF_{i,j},$$
(1)

where:

(*Intake*)_{*j*,*p*}(*t*) = intake rate of radionuclide j at time t (Bq/yr or pCi/yr), *M* = the number of initially existent radionuclides, $ETF_{j,p}(t)$ = environmental transport factor for radionuclide j at time t (g/yr), *p* = primary index of pathway, $SF_{ij}(t)$ = source factor, *i*,*j* = index of radionuclide (i for the initially existent radionuclide and j for the radionuclides in the decay chain of radionuclide i), $S_i(0)$ = initial soil concentration of radionuclide i at time 0,

The cancer risk at a certain time point from external exposure can be estimated directly by using the risk coefficients, which are the excess cancer risks per year of exposure per unit of soil concentration. Because the risk coefficients are derived on

the basis of the assumptions that the contamination source is infinite both in depth and lateral extension and that there is no cover material on top of the contaminated soil, it is necessary to modify the risk coefficients with the cover and depth, shape, and area factors to reflect the actual conditions. Non-continuous exposure throughout a year also requires that the occupancy factor be considered when calculating the cancer risks.

Consequently, the RESRAD code uses the environmental transport factor for the external radiation pathway, along with the risk coefficient and exposure duration, to calculate the excess cancer risks as seen in Equation (2) [6]:

$$(Cancerrisk)_{j,1}(t) = \sum_{i=1}^{M} ETF_{j,1}(t) \times SF_{ij}(t) \times S_{i}(O) \times BRF_{i,j} \times RC_{j,1} \times ED, \quad (2)$$

where:
$$RC_{j,1} = \text{risk coefficient for external radiation (risk/yr)/(pCi/g),}$$

ED = exposure duration (30 yr).

For the inhalation and ingestion pathways, the cancer risks at a certain time point are calculated as the products of intake rates, risk coefficients, and exposure duration. Unlike the intake rates, cancer risks from inhalation of radon and its decay progeny are reported as total risks that include radon and progeny contributions. Therefore, the radon risks are the sums of the products of the individual radon or progeny intake rates and their risk coefficients.

4.4. Model Parameters

The model for assessing the dose rates and risks arising from hot-cells clean-up activities in the decommissioning of the VVR-S Research Reactor is based on a previous work described in Reference [7]. The input parameters of the model are given in Table 5. The external exposure of the worker due to floor contamination taking into considering the following assumptions: 1 compartment - Room 43; source geometry – area (circle with the 1 cm radius); 10 distinct source locations (the 10 most significant sources locations); 10 receptor locations and 1 decontamination cycle, 120 min long. The calculation was performed over 5-time intervals (initially, after 1 day, after 3 days, after 7 days and after 10 days (the end of the cleaning process)) during 10 days period of cleaning operations. For modeling purpose, a room sketch was considered to be used in the ResRad Build model (see Fig.10).

Parameter	Unit	Building Renovation	Remarks
Exposure duration	days (d)	10.00	The total length of time considered by the dose assessment, including intervals during which receptors may be absent from the building or a contaminated indoor location
Indoor fraction	dimensionless	0.003	Fraction of the exposure duration spent by one or more receptors inside a building (working time/exposure duration)
Receptor location			Position relative to source centre
Receptor inhalation rate	m ³ /d	28.8 [9]	For building renovation scenario considering a 1.6 m ³ /h breathing rate of moderate activity given in the EPA Exposure Factor Handbook (EPA 1997) in 24 hours
Receptor indirect ingestion rate	m²/h	0.00001 [9]	Due to the mask wearing
Source type -	_	Area	Source geometry
Direct ingestion rate	1/h (area) g/h (volume)	0.052 [9]	Calculated from the default ingestion rate of 1.1×10^{-4} m ² /h in NUREG/CR-5512 building occupancy scenario (Beyeler et al. 1999 [9]). The effective transfer rate from NUREG/CR-5512 building renovation scenario for ingestion of loose dust to the hands and mouth during building renovation (Wernig et al. 1999 [9]).
Air release fraction	_	0.0001 [9]	Fraction of mechanically removed or eroded material that becomes airborne. For the building renovation scenario, a smaller fraction is breathable.
Removable fraction	-	Not Required for current analysis [10]	10% of the contamination is removable (NUREG/CR- 5512 building occupancy scenario default [10]). The parameter is not required for the volume source.
Time for source removal or source lifetime	d	Not Required for current analysis [10]	Value for the building occupancy scenario is the most likely value from the parameter distribution. The parameter is not required for the volume source.
Source erosion rate	cm/d	0 [10]	Due to short time of decontamination process

Table 5) Model input parameters [7] [8]

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5. Results

External

The results showed a low dose and risk associated with the clean-up and decontamination activities of the depleted uranium workshop. The received dose rate was 2.25 mSv/h at the beginning and decreased to 0.7 mSv/h after one cycle of decontamination (see Fig. 11). The afferent cancer risk for the worker was lower than 1.80×10^{-4} (see Fig. 12). The maximum exposure is due to the inhalation of the dust particle. It was assumed that after a second decontamination cycle using DeconGel the dose would fall to background levels. Therefore, a two-cycle decontamination procedure was recommended for the whole uranium workshop (rooms 43-47). The dose was still higher than background probably due to particle resuspension in the air.



📕 Inhalation 📕 Deposition 🦊 Immersion 📕 Ingestion

Fig. 11. Dose rate received by worker by time and pathway for summed nuclides





6. Conclusions

In case of the evaluated dose, most of the contribution came first from the inhalation pathway and second from external pathway at contact. This imposed a conservative designing approach for the clean-up operations (2 decontamination cycles, rotating workers, minimizing time inside) and the use of enhanced radiological equipment (respiratory filters, complete full body working suits and one time use gloves). All these measures contributed to the calculated extremely low cancer risk.

The methodology presented can be applied for a more complex modelling of the whole processing workshop including also rooms 44-47, of which room 44 presented the main challenge being the most contaminated, as revealed by the measurements performed during its clean-up in 2012.

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REFERENCES

- [1] International Atomic Energy Agency, Topics/Spent Fuel Management/Depleted Uranium.
- [2] Depleted uranium, sources, exposures and health effects, Department of Protection of the Human Environment, World Health Organization, Geneva, (2001).
- [3] D. Stanga, E. Ionescu, D. Radu, Proiectul de Dezafectare al Reactorului Nuclear VVR-S IFIN-HH - Document suport - Raport de Caracterizare Radiologica, Bucuresti, Magurele, (2010).
- [4] C. Yu, D.J. LePoire, J.J. Cheng, E. Gnanapragasam, S. Kamboj, J. Arnish, B.M. Biwer, A.J. Zielen, W.A. Williams, A. Wallo III, H.T. Peterson, Jr., User's Manual for RESRAD-BUILD Version 3, Environmental Assessment Division, Argonne National Laboratory (ANL), ANL/EAD/03-1, (2003).
- [5] U.S. Environmental Protection Agency, Exposure Factor Handbook, EPA/600/P-95/002Fa, Office of Research and Development, National Center for Environmental Assessment, (1997).
- [6] K.F. Eckerman, et al., Cancer Risk Coefficients for Environmental Exposure to Radionuclides, EPA 402-R-99-001, Federal Guidance Report No. 13, prepared by Oak Ridge National Laboratory, Oak Ridge, Tenn., for U.S. Environmental Protection Agency, Office of Radiation and Indoor Air, (1999).
- [7] A. O. Pavelescu, V. Popa, M. Dragusin, Modelling of the Dose Rates and Risks Arising from Hot-cells Clean-up Activities in the Decommissioning of the VVR-S Research Reactor, Romanian Reports in Physics, Year 2012, Vol. 64, No. 2, (2012).
- [8] C. Tuca, A. O. Pavelescu, Dose assessment in decontamination process of the Hot Cells from VVR-S Nuclear Research Reactor, Seventh International Conference on Radiation in Various Fields of Research (RAD7), Herceg Novi, Montenegro, 10-14 June, (2019).
- [9] W.E. Beyeler, et al., Residual Radioactive Contamination from Decommissioning; Parameter Analysis, NUREG/CR-5512, Vol. 3, Nuclear Regulatory Commission, Office of Nuclear Regulatory Research, October, (1999).
- [10] B.M Biwer, et al., Technical Basis for Calculating Radiation Doses for the Building Occupancy Scenario Using the Probabilistic RESRAD-BUILD 3.0 Code, NUREG/CR-6755, ANL/EAD/TM/02-1, Argonne National Laboratory, Office of Nuclear Regulatory Research, (2002).