DOI: 10.5604/20830157.1130170

DECONTAMINATION OF CONTAMINATED SCRAP METAL BY MELTING ARISEN FROM THE DECOMMISSIONING OF NUCLEAR POWER PLANT SHUT DOWN AFTER AN ACCIDENT

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Abstract. *The paper briefly describes the decontamination of radioactive scrap metal by melting as prospective technology to reduce the amount of metallic radioactive waste. Decontamination by melting presents a particular advantage of homogenising a number of radionuclides in the ingots and concentrating other radionuclides in the slag and dust filter resulting from the melting process, thus decontaminating the primary material. Presented paper also deals with the evaluation of an exposure of the workers during melting of radioactive scrap metal as well as impact of melting on the environment. According to obtained results, the effective doses absorbed by workers during the melting meets legislatively given limit 20 mSv annually.*

Keywords: metal melting, metallic radioactive material, VISIPLAN 3D ALARA, exposure

PRZETAPIANIE JAKO METODA DEKONTAMINACJI SKAŻONEGO ZŁOMU Z ROZBIÓRKI ELEKTROWNI ATOMOWEJ ZAMKNIĘTEJ PO AWARII

Streszczenie. *W artykule krótko opisano dekontaminację radioaktywnego złomu poprzez przetapianie, jako potencjalną technologię do zmniejszenia ilości metalowych odpadów radioaktywnych. Szczególną zaletą dekontaminacji poprzez przetapianie jest homogenizacja szeregu radionuklidów we wlewkach i zatężenie innych radionuklidów w żużlu i na filtrze pyłu powstających podczas procesu przetapiania, a tym samym dekontaminacji materiału podstawowego. Prezentowana praca dotyczy również oceny ekspozycji pracowników podczas przetapiania radioaktywnego złomu, jak również wpływu przetapiania na środowisko. Według uzyskanych wyników, skuteczne dawki pochłaniane przez pracowników podczas przetapiania spełniają ustawowy limit 20 mSv rocznie.*

Słowa kluczowe: przetapianie metalu, metaliczny materiał promieniotwórczy, VISIPLAN 3D ALARA, ekspozycja

Introduction

At present there are more than 400 nuclear power plants (NPP) and many other nuclear installations (NI) in the operation worldwide. However, everyone technological equipment is limited by its lifetime and after its expiration the nuclear power plant (or NI) has to be decommissioned and dismantled. It is estimated by the OECD's Nuclear Energy Agency that about 400 commercial nuclear power plants will be decommissioned till 2050, which may result in more than 5 million tonnes of scrap metal suitable for recycling. Considering all other types of NI that will also be decommissioned, the amount of scrap metal arisen from decommissioning in the coming decades is estimated to be about 30 million tonnes [4].

The large amount of slightly radioactive scrap metal arising from decommissioning of NI may present notable problems in radioactive waste management. Currently operating waste disposal facilities cannot accommodate such large volumes of metal waste (for reasons of costs or because of public opposition to expansion of available waste capacity or to the sitting of new disposal capacities) and therefore decontamination and recycling is a suitable way to reduce significant amounts of waste.

1. Decontamination by melting

Metal melting is a thermal desorption process, during which different elements and their radioactive isotopes are redistributed between the slag, the metal, and the off-gases depending on elemental properties. The more volatile elements (including their radioactive isotopes) – such as caesium, iodine or hydrogen (tritium) – leave the melt and are essentially transferred to the off-gases or, in some cases, to the slag. Some metallic elements (including radioactive isotopes) – such as cobalt, nickel, chromium, iron, zinc and manganese – mainly remain within the melt. Transuranic elements can be readily oxidized and will transfer to the slag [2].

A particularly advantageous consequence of melting is its "decontamination" effect on ^{137}Cs, a volatile element that has a half-life of 30 years. During melting, ^{137}Cs accumulates in the dust collected by ventilation filters and is removed. The dominant remaining nuclide in the ingots (for most reactor scrap) is ^{60}Co. This element has a half-life of only 5.3 years. Other remaining nuclides have even shorter half-lives. Consequently, ingots with reasonably low-activity concentrations may be stored for release in a foreseeable future [5].

The final product (ingot, shielding block, centrifugated steel cylinder, etc.) is homogeneous, stable, and has the remaining activity content bound in the metal. Melting can produce a conditioned waste form suitable for direct disposal. Metal melting provides several advantages such as:

- Extensively proven technology.
- High volume reduction. If recycling is possible, the volume reduction factor from the disposal perspective of up to 100 is possible.
- The end product is typically homogenous and stable with remaining activity content bound in the metal.
- The end product can be reused and recycled within nuclear or conventional metal industry.

2. The relevance of metal melting in Slovakia

Two nuclear power plants are being decommissioned in Jaslovské Bohunice, Slovakia. One is A1 NPP (heavy water gas cooled reactor) shut down in 1977 after an accident with local consequences (level 4 in the INES scale) after five years of operation. The decommissioning of this NPP has started in 1998 and should end late in 2033. Its decommissioning is very difficult due to the high contamination of primary circuit. The second NPP in decommissioning process is V1 NPP (Russian type of pressurized water reactor, VVER-440 twin unit) shut down after 28 years of standard operation. The decommissioning of V1 NPP has started in 2011 and should end in 2025. Both NPPs present potential source of large amount of low level metallic radioactive waste suitable for decontamination and recycling. As was mentioned earlier, metal melting is suitable decontamination technology that could be helpful during the decommissioning of mentioned NPPs. At the present no melting unit is commissioned in Slovakia but it is planned to construct and commission such facility in the near future. Application of metal melting in the waste management strategy could lead to releasing considerable volume of scrap metal arisen from both Slovak NPPs that are currently being decommissioned and also from NPPs that will be decommissioned in the future. Melting technology can be also considered for volume reduction prior disposal.

3. Radiation impact on the workers during the melting

Metal melting is a complex process starting by delivery of radioactive scrap metal to the melting facility, ending by releasing of metal ingots to the environment to restricted or unrestricted reuse. As was mentioned before, the paper deals with the evaluation of an exposure of the several workers, specifically scrap truck driver, scrap cutter, furnace operator, slag remover, ingot caster, ingot handler.

3.1. General assumptions of model calculation

In the calculation of radiation impacts of selected workers following considerations were taken into account:

- Melting facility comprises induction furnace of charge size of approx. 2 tonnes of scrap metal.
- The melting facility is able to melt two batches per one workday, i.e. approx. 2 tonnes of scrap metal.
- 250 workdays per year are considered, i.e. the annual capacity of melting facility is approx. 1 000 tonnes of scrap metal.
- Radiological limitation for the facility is 1 000 Bq/g for total β/γ activity (it is conservatively considered that workers melt entire year scrap metal with maximum allowed activity).
- Considered exposure pathways are external as well as internal.
- For the calculation one nuclide vector is used. This nuclide vector characterizes radiological situation of NPP shut down after fuel accident (A1 NPP). Its composition is shown in Table 1.
- During the melting radioactivity distribution coefficients for each radionuclide is considered. It is necessary to know the fraction of the activity originally present in radioactive scrap metal which may be transferred to the ingot, to the slag and to the dust after melting. These coefficients were adopted from the experiences obtained in the CARLA Plant, Germany [7] and from publication "NUREG-1640" [10] and are shown in Table 1.
- Mass partitioning factors for the melt, slag and dust are also considered (98.35% for melt/ingot, 1.64% for slag and 0.01% for filter dust) [9].

Table 1. Nuclide vector composition and radioactivity distribution coefficients during melting used in calculations

Radionuclide	Share [%]	Half-life [yr]	Distribution [%]		
			Melt	Slag	Dust
^{137}Cs	48.41	30	1	60	39
^{90}Sr	32.66	28.70	1	97	2
^{63}Ni	13.15	100	90	10	0
^{60}Co	2.77	5.27	88	11	1
^{241}Am	1.28	432	1	97	2
^{152}Eu	0.75	13.50	4	95	1
^{239}Pu	0.60	24100	1	97	2
^{151}Sm	0.37	90	0	93	7

3.2. General description of worker scenarios

For dose assessment purposes, six representative worker scenarios for metal melting were developed. In the following paragraphs the brief description of selected workers is given.

The <u>*truck driver*</u> scenario models the potential dose to a worker who transport contaminated scrap metal from the place of its origin to the melting facility. It is estimated that one worker would spend approx. one and half hour driving the truck to the melting facility. Considering that scrap metal is transported in ISO container with maximum load of 28 tonnes, the worker would perform 36 transports to transport 1 000 tonnes of contaminated scrap metal annually. Because the melting facility and decommissioned NPPs are in the same locality the truck driver scenario is relevant only for metallic radioactive waste arisen outside Jaslovské Bohunice locality.

The <u>*scrap cutter*</u> scenario models a worker who prepares the scrap for delivery to the furnace. This worker´s activity includes shredding, cutting and sorting of scrap metal. It is estimated that the worker would have to work 6 hours per day to process approx. 4 tonnes of scrap metal.

The <u>*furnace operator*</u> scenario models the potential dose to a worker who operates furnace in furnace control room. It is considered that this worker, besides operating the furnace during the melting, also operates the crane and loads scrap metal into the furnace. It is estimated that the worker have to work approx. 3.5 hours per day to load and melt two batches, i.e. 4 tonnes of scrap metal.

The <u>*slag remover*</u> scenario models the potential dose to a worker who uses standard loading and unloading equipment to handle the slag by-product at the melting facility. In general slag can be removed in several different ways, i.e. removed from the top of the furnace manually or by manipulator (crane), casted into ingot ladle and removed after cooling. In the calculations is assumed that the worker removes the slag manually from the melt bath surface. It is estimated that the worker would have to work 1.25 hours per day to process the slag from two batches.

The <u>*ingot caster*</u> scenario models a worker casting metal ingots. The melt is casted into 400 kg ingot moulds. It is estimated that the worker would have to work 2 hours per day to cast 10 ingots.

The <u>*ingot handler*</u> scenario models the potential dose to a worker manipulating the ingots. This worker's activity includes pulling out the ingots of the ingot moulds and its replacement to 200 L drums. The ingots are measured in gamma scanner for residual activity and subsequently free released or transported to interim storage facility. It is estimated that the worker manipulates the ingots approx. 1.5 hour daily

3.3. Radiological impact assessment method

The main purpose of the paper is to assess radiation impacts of the workers during the melting of contaminated scrap metal. Generic radiation exposure scenarios were used to conceptually model situations regarding melting. These scenarios are a combination of radiation exposure pathways containing specific exposure conditions. Three main exposure pathways are considered in calculations, exposure to external radiation, inhalation of radioactive small particles or gases and ingestion of radioactive materials. In following chapters formulas for the assessment of particular exposure pathways are described [1, 3]. Calculation of external exposure was performed using the computational tool VISIPLAN 3D ALARA Planning Tool.

External exposure (VISIPLAN 3D ALARA description)

VISIPLAN, developed in Belgium, is an appropriate tool for evaluation of an external gamma and x-ray exposure. This tool allows modelling of real scenarios hence the obtained results can be beneficial for nuclear management practices. Using this tool, decommissioners can model from simple up to complex geometries, thus providing reliable results. VISIPLAN can be also used for radiation protection purposes in nuclear installations decommissioning and waste management strategy.

The method used in VISIPLAN is based on a point-kernel calculation with a build-up correction, where the volume source is divided into point sources. The photon fluence rate at a dose point is then determined by superposition of partial dose contributions from single point sources:

$$\phi = \int_V \frac{SBe^{-b}}{4\pi r^2} dV \qquad (1)$$

where: S – source strength per unit volume [n/s], B – build-up factor [-], b – dimensionless term which represents the attenuation effectiveness of the shield, r– distance from the point source [m], V – volume [m^3].

Point sources are called "kernels" and the process of integration, where the contributions to the dose from each point is added up, is called a "point-kernel" method [11].

Inhalation

For the inhalation exposure, following formula was used [3]:

$$E_{inh,C} = e_{inh} t_e f_d f_c C_{dust} \dot{V} e^{-\lambda t_1} \frac{1-e^{-\lambda t_2}}{\lambda t_2} \quad (2)$$

where: $E_{inh,C}$ – committed effective dose in a year from inhalation per unit activity concentration in the material [(μSv/a)/(Bq/g)], e_{inh} – effective dose coefficient for inhalation [μSv/Bq], t_e – exposure time [h/a], f_d – dilution factor [-], f_c – concentration factor of specific activity in the fine fraction [-], C_{dust} – effective dust concentration in the air [g/m^3], $\dot{V}$ – breathing rate [m^3/h], λ – radioactive decay constant [1/a], t_1 – decay time before the start of the scenario [a], t_2 – decay time during the scenario [a].

Ingestion

For the ingestion exposure, following formula was used [3]:

$$E_{ing,C} = e_{ing} q f_d f_c f_t e^{-\lambda t_1} \frac{1-e^{-\lambda t_2}}{\lambda t_2} \quad (3)$$

where: $E_{ing,C}$ – committed effective dose in a year from ingestion per unit activity concentration in the material [(μSv/a)/(Bq/g)], e_{ing} – effective dose coefficient for ingestion [μSv/Bq], q –ingested quantity per year [g/a], f_d – dilution factor [-], f_c – concentration factor in the fine fraction [-], f_t – root transfer factor [-], λ – radioactive decay constant [1/a], t_1 – decay time before the start of the scenario [a], t_2 – decay time during the scenario [a].

3.4. Model description and definition of input parameters used in calculation

Following chapter deals with the description of model and definition of general parameters used in radiation impact assessment. In Table 5 general parameters for melting of contaminated scrap metal are shown.

Table 2. General parameters used in model calculation

Worker scenario	Radiation source	Distance of worker from the source [m]	Annual exposure time [h]
Truck driver	Scrap metal	3.5	54
Scrap cutter	Scrap metal	0.5 – 5	1 500
Furnace operator	Scrap metal, molten metal, slag	3 – 4	875
Slag remover	Slag, molten metal	0.5 – 1	315
Ingot caster	Molten metal	0.75 – 1.25	500
Ingot handler	Ingot	0.5 – 1	375

During the transport of scrap metal, the internal exposure pathway is excluded, because a driver would not be directly in contact with the radioactive scrap metal, thus the only pathway considered is external exposure. For the external dose calculation, the source is modelled as a 28 tonnes rectangular-shaped box placed in ISO container with a density of 2.5 g/cm^3.

The scrap cutter scenario considers external exposure, inhalation and ingestion of airborne particles from the cutting process. For the external exposure purposes the source is modelled as a two boxes full of scrap metal with a density of 2.5 g/cm^3 and weight of approx. 4 500 tonnes and piece of scrap metal (tube) that is being fragmented.

The exposure pathway evaluated for the furnace operator scenario is only external exposure, because it is considered that furnace control room is separate closed object and no dust and airborne particles can penetrate into it. The source of the radiation during the loading of scrap metal into the furnace represents the box filled with the metal and partially loaded furnace. The source during the melting represents molten metal within the furnace. The source during the removing of the slag represents molten iron and slag at the top of the furnace and the source during casting the ingots represents decontaminated molten metal. The furnace is modelled as tube with an internal diameter of 0.7 meter and height of 1.2 meter and the source of the radiation (molten metal) is placed within the furnace. The thickness of the furnace wall is 0.1 meter.

The slag remover scenario includes only removal of the slag from the furnace and its replacement to the area, where radioactive waste (by-products) is stored. The exposure pathways considered for this scenario include external as well as internal (inhalation and ingestion) exposure. For external exposure purposes is the slag modelled as a 40 kg cylinder with density 3 g/cm^3.

The ingot caster scenario considers external exposure, inhalation and ingestion pathways. The ingot is modelled as 400 kg block (cylindrical) with diameter and height of 0.4 meter. The source of radiation for this scenario is considered molten metal casted in three ingot moulds and the melt remaining in the furnace after casting mentioned three ingots (total mass of the radiation source is the mass of one batch after removal of the slag).

The radiation source in ingot handler scenario is considered ingots resulting from two melt (10 ingots). Considered exposure pathways are external as well as internal exposure.

In the table 3 and table 4 input parameters used for calculation of internal exposure are shown.

Table 3. Input parameters used for calculation of an internal exposure [1, 3]

Inhalation		
C_{dust} [g/m^3]	f_d [-]	V [m^3/h]
5.00E-04 for fragmentation	0.1	1.2
1.00E-03 for other scenarios		
Ingestion		
q [g/h]	f_d [-]	f_c [-]
5.00E-3	0.1	2

Table 4. Effective dose coefficient for inhgalation and ingestion and concentration factor for inhalation [1, 3]

Radionuclide	e_{inh} [Sv/Bq]	e_{ing} [Sv/Bq]	Concentration factor for inhalation f_c [-]
^{137}Cs	6,70E-09	1,30E-08	70
^{90}Sr	1,51E-07	3,07E-08	10
^{63}Ni	4,40E-10	1,50E-10	1
^{60}Co	9,60E-09	3,40E-09	1
^{241}Am	3,90E-05	2,00E-07	10
^{152}Eu	3,90E-08	1,40E-09	10
^{239}Pu	4,70E-05	2,50E-07	10
^{151}Sm	3,70E-09	9,80E-11	10

3.5. Obtained results

The results presented in this paper are based on generic exposure scenarios and pathways analyses using one nuclide vector composition, which represents radiological situation of NPP shut down after fuel accident.

Obtained results for described worker scenarios are shown in Table 5. As one can see from the results, the absorbed effective dose depends on several factors like dominant radionuclides in nuclide vector, radioactivity distribution during the melting, time of performed activity, etc.

Table 5. Obtained results for melting of scrap metal

	Absorbed effective dose [mSv/a]			
	Inhalation	**Ingestion**	**External exposure**	**Sum**
Truck driver	-	-	1.66E-01	**1.66E-01**
Scrap cutter	7.90E-02	1.45E-02	7.50E+00	**7.59E+00**
Furnace operator	-	-	1.28E+00	**1.28E+00**
Slag remover	1.87E+00	1.56E-01	6.50E+00	**8.53E+00**
Ingot caster	5.37E-04	7.12E-05	2.00E-01	**2.01E-01**
Ingot handler	4.03E-04	2.37E-05	8.50E-02	**8.54E-02**

According to the obtained results, the slag remover receives the highest dose, because he is working with the secondary radioactive waste with relatively high concentrations of several radionuclides. As mentioned in the chapter describing the melting technology, decontamination effect (factor) is much higher for fission products as well as transuraniums, which are mostly redistributed to the slag. The lowest dose is received by worker during the ingots manipulations, because the ingot is decontaminated and the residual activity is caused only by low concentrations of activation products.

In general it can be said that external exposure in comparison with internal exposure is dominant, only exception is removing the slag, because the slag remover works with secondary RAW with high concentrations of fission products as well as actinides.

VISIPLAN 3D ALARA code also allows graphical representations of obtained results. Some of these figures can be seen below (Fig 1. and Fig 2.).

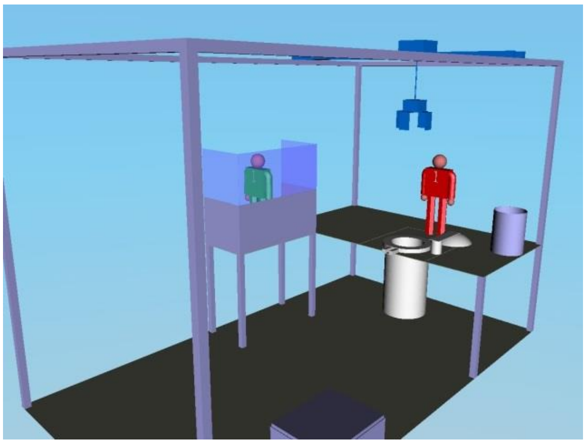

Fig 1. Furnace operator and slag remover modelled in VISIPLAN 3D ALARA

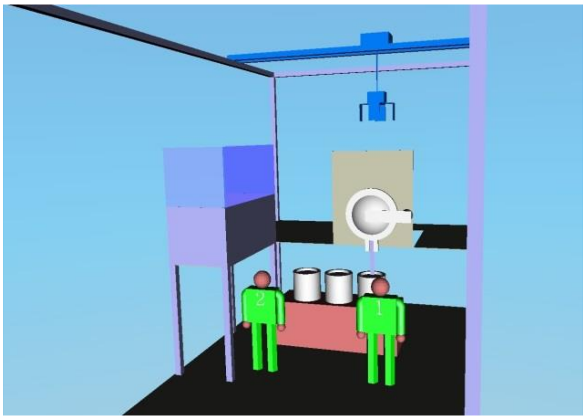

Fig 2. Workers during casting the ingots modelled in VISIPLAN 3D ALARA

Apart from figures depicted above, VISIPLAN 3D ALARA also provides a graphical output in the form of radiation fields and radiation maps at any distance from the source. Following figures depict radiation situation (dose maps) in working areas during different activities in the melting process. All dose maps are calculated at height of 1.2 m above the floor. Fig. 3 depicts radiation satiation during the fragmentation and sorting of scrap metal. Fig. 4 depicts radiation situation during the melting, when the furnace is fully loaded and closed and Fig. 5 depicts dose map during casting the ingots.

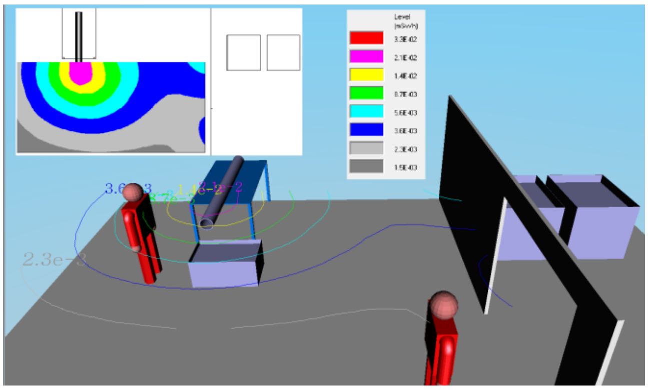

Fig 3. Dose map during the fragmentation activities modelled in VISIPLAN 3D ALARA

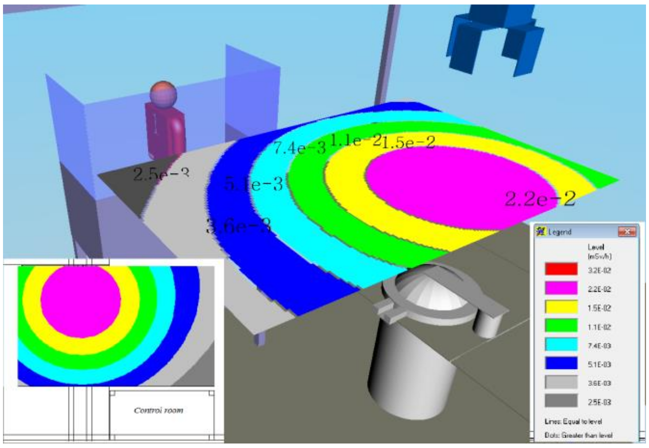

Fig 4. Radiation situation during melting modelled in VISIPLAN 3D ALARA

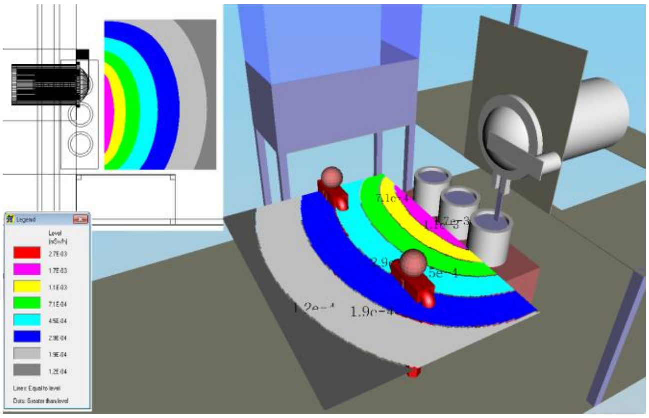

Fig 5. Dose map during casting the ingots modelled in VISIPLAN 3D ALARA

4. Radiation impact on the environment

The melting facility presents a source of discharges into the environment occurring during the melting of contaminated scrap metal. Therefore the ventilation and filter system is one of the most important safety elements of such facility. At the present metallic radioactive waste is treated at fragmentation and decontamination facility located in Jaslovské Bohunice locality, with its own central filtration and ventilation system. This facility should be extended and the melting furnace should be part of the facility in the near future. The furnace area should have its own independent filtration and ventilation system and air from the furnace cleared by this system will be transferred to the central filtration system. Such combination of two filtration systems must assure that discharge of radioactive material (dust) into the environment does not exceed limit values stated by Public Health Authority of Slovak Republic. It is expected that total filtration efficiency will be at least 99.95%. As example melting plant CARLA in Germany can be given, where filtration system is achieving efficiency of 99.997% [6]. Considering the efficiency of 99.997% only approx. one percent of permitted limits would be discharged into the environment.

5. Conclusion and discussion

The main goal of the paper is to evaluate radiation impact on the workers as well as on the environment during the melting of contaminated scrap metal. As can be seen from the obtained results, the worker´s received dose depends on the performing activity as well as on radionuclide present in the scrap metal as contaminant and radioactivity distribution coefficients during melting. The highest dose is absorbed by slag worker (8.53 mSv annually), because of manual manipulation with the slag. The impact on the environment is not so significant because only small fraction of annual limits is discharged considering filtration and ventilation efficiency similar to the CARLA plant.

Generally it can be said that external exposure pathways is much more relevant as internal exposure pathway. The workers in the melting facility wear protective suits during melting and preparation works resulting in minimal ingested or inhaled dust.

It is important to note that evaluated worker scenarios are basic tasks during melting. The complex assessment of metal melting process requires the evaluation of all activities related to melting, e.g. change of used furnace lining, manipulation with collected dust from ventilation system, transport of ingots to the ingot storage yard, etc.

Mentioned values (absorbed effective doses) meet legislatively given limits in Slovak Republic, in which value of 20 mSv [8] is defined as the maximum allowed dose received by a worker annually. It is also important to note that doses are calculated conservatively, because it is unlikely that the workers would be entire year melting the scrap metal with limit values (1 000 Bq/g).

It is also important to note that presented paper serves as preliminary study and real individual effective doses received during the melting of scrap metal in Slovakia can be quite different because some general assumptions taken into account during calculation were adopted from experiences obtained in the melting facilities in the world. Specifically distribution coefficients during the melting was adopted from CARLA plant experiences, but planned melting plant in Slovakia is similar (induction furnace with quite lower charge size), therefore the distribution coefficients are likely to be very similar. The same can be said about the filtration system.

6. Acknowledgement

This project has been partially supported by the Slovak Grant Agency for Science through grant VEGA 1/0796/13 and by the Ministry of Education by decree CD-2009-36909/39460-1:11 within the bounds of project CONRELMAT.

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otrzymano/received: 2013.09.28 *przyjęto do druku/accepted: 2014.11.10*