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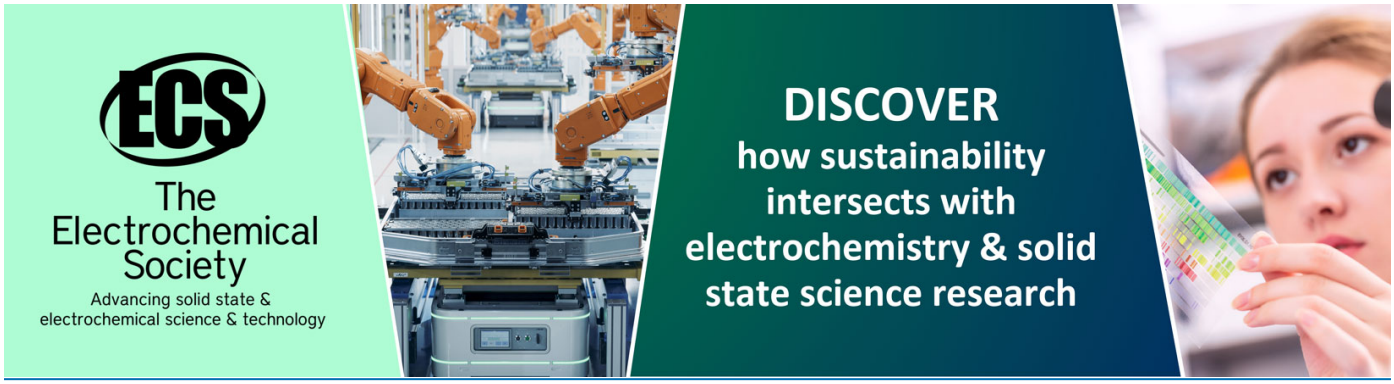

IOP Conf. Series: Journal of Physics: Conf. Series **1115** (2018) 052024 doi:10.1088/1742-6596/1115/5/052024

Activity of irradiated beryllium and waste management after its purification

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Abstract. The relevance of work is determined by the fact that development of irradiated beryllium recycling method in nuclear industry is environmentally and economically proven step. Currently the feasibility study to develop such method is carried out in the IAE RSE NNC RK. Determination of beryllium activity spent in nuclear power facilities will allow to evaluate and define conditions for studying of irradiated beryllium. Activity calculations of beryllium activation samples and impurities irradiated in JMTR reactor for several years have been carried out. The dose rate of gamma irradiation from irradiated beryllium sample at work places close to a plant, operation time at these places and permissible doses to the personnel were determined. Calculation results will be used at preparation for reprocessing of irradiated beryllium and for future RW management resulted from purification. During operation of the plant for purifying irradiated beryllium, liquid and solid RW will be generated. The paper provides the methods of RW management.

1. Introduction

Beryllium holds a special place in nuclear industry, since this metal is a structural material of some reactors and ampoule neutron sources [1-4]. Beryllium is characterized by a high radiation resistance continued under high temperature, small section of neutron capture and large section of their dispersion. Beryllium enables operating the thermal neutron reactors and performing experiments to produce controlled fusion power – energy for future. This is the only nature material, which capable maintain fusion reaction without contamination of high temperature plasma, where it occurs [5-9].

Reprocessing of irradiated beryllium for its reusing will allow significantly advance environmental safety of nuclear industry and profitability of nuclear power plants.

This works perform estimation of experimental research conditions at the plant for purifying of irradiated beryllium. The method is consist in conversion of beryllium and its radionuclides into chlorides, which leads to the generation of liquid, gas and solid RW [10-12]. The sample for research is beryllium cylinder-shaped reflector extracted from the JMTR reactor (figure 1), which was divided into 10 parts.

2. Determination of beryllium sample activity and dose rate values

In order to evaluate current activity of irradiated sample, neutronic calculations have been carried out. Beryllium was irradiated since 1968 till 1975, therewith energy release in the reactor was 24017.4

MWD. For JMTR reactor power of 50 MW, thermal neutron density flux is approximately equal to $8.0 \cdot 10^{13}$ n/cm^2/s and respectively, fast neutron density ~$7.5 \cdot 10^{12}$ n/cm^2/s.
The activity of impurities in beryllium samples was calculated by the formula (1):

$$A(t) = A_0 \cdot \exp(-0{,}693 \cdot t/T_{1/2}) \quad (1)$$

where A_0 – initial activity as of 1 July 2009;
$A(t)$ – calculation of activity value as of 1 July 2018.

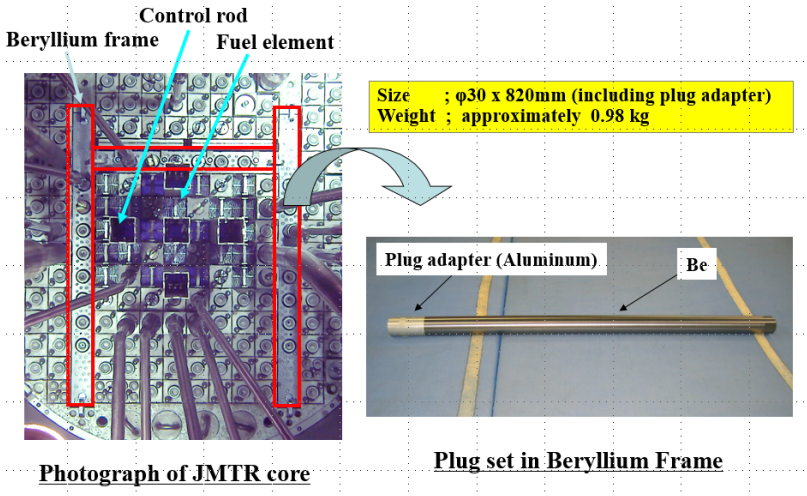

Figure 1. Irradiated Beryllium Plug in JMTR Core.

Table 1 provides half-decay periods, initial activity A_0 of the most active beryllium block (block No. 3) as of 1 July 2009 and calculation of activity values $A(t)$ as of 1 July 2018.

Table 1. Half-decay periods, initial activity of radioactive impurities of beryllium block as of 1 July 2018.

Isotope	$T_{1/2}$ (year)	A_0 (Bq)	A(t) (Bq)
^{3}H	12.3	$2.961 \cdot 10^{10}$	$1.81 \cdot 10^{10}$
^{10}Be	$1.601 \cdot 10^{6}$	$3.441 \cdot 10^{6}$	$3.41 \cdot 10^{6}$
^{55}Fe	2.73	$5.331 \cdot 10^{5}$	$5.41 \cdot 10^{4}$
^{59}Ni	75000	$4.391 \cdot 10^{5}$	$4.41 \cdot 10^{5}$
^{60}Co	5.27	$3.201 \cdot 10^{7}$	$9.81 \cdot 10^{6}$
^{108m}Ag	127	$2.841 \cdot 10^{6}$	$2.71 \cdot 10^{6}$

Table 2. Results of measurements and calculation of specific activity of radioactive impurities as of July 2018.

Isotope	$T_{1/2}$ (year)	A_0 (Bq)	$A(t)$ (Bq)
^{60}Co	5.27	$2.541 \cdot 10^{4}$	$1.01 \cdot 10^{4}$
^{108m}Ag	127	$1.11 \cdot 10^{3}$	$1.11 \cdot 10^{3}$

In 2011, specific activity measuring of beryllium samples have been carried out. Table 2 provides results of measurements (A_0) and specific activity calculation of radioactive impurities as of July 2018 ($A(t)$).

The dose rate of the sample at different distance has been calculated. When calculating, the following radiation intensity was taken (calculated values as of 1 July 2018):

- gamma-radiation of ^{60}Co isotope, quantum /s $1.96 \cdot 10^7$
- β- radiation of ^{60}Co isotope, freq./s $9.80 \cdot 10^6$
- β- radiation of ^{3}H isotope, freq./s $1.80 \cdot 10^{10}$

Radiation shielding was not considered during calculations. The calculation were performed using two independent methods: with special program and by analytical method. To calculate dose rate distribution of photon and electron radiation, MCNP5 calculation code was used with a library of photon and electron constants from the ENDF/B-VI and el03 libraries [13]. All the initial data, assumptions and terms were taken on the basis of the conservative calculation principle, i.e. any deviation of the calculation model from the real conditions should lead either to an increase in the calculated doses received by the personnel or not to affect them significantly (within the error of the calculations).

The following assumptions and simplifications were made during dose rate calculation of outer irradiation using analytical method:

- In calculations, self-absorption of ionizing radiation in the source material was neglected, i.e. the radiation source was considered as a point source;
- Only the contribution from the ^{60}Co isotope was taken into account.

In the analytical method, the exposure dose rate for a given calculated geometry was determined by the formula [14-17]:

$$P = P_\gamma \times A/R^2, \quad (2)$$

wherein

P – exposure dose rate, R/h; R – distance from the source to measure point, cm; A – activity, mCi; P_γ – the total gamma constant of the ^{60}Co isotope, 12,93 R*cm^2/h*mCi [24].

The equivalent dose rate of ionizing radiation in air was determined by the formula:

$$Q = k \times P, \quad (3)$$

where in Q – equivalent dose rate, Sv/h; P – exposition dose rate, R/h; k – conversion factor for air.

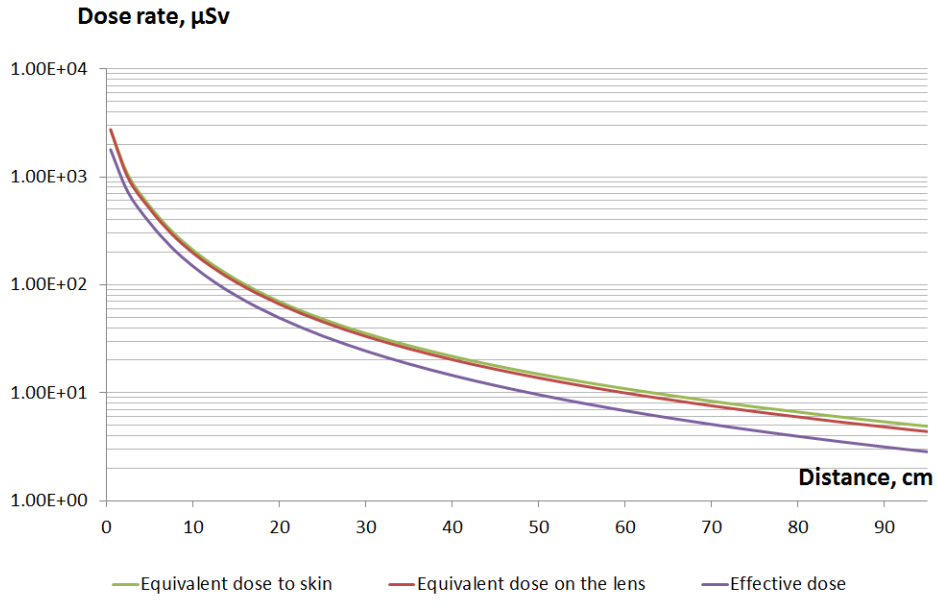

Figure 2. The values of the effective and equivalent dose rate from the ionizing radiation source.

Then, comparison and analysis of the calculation results have been carried out. As a result, data were obtained on the dose fields of electron and photon radiation from a source of ionizing radiation. The main contribution to the dose rate is made by the gamma radiation of the ^{60}Co isotope.
The doses from the ionizing radiation source for the area up to 1 m were calculated. The results of calculating the effective dose and the equivalent dose on the skin and on the lens from ionizing radiation, obtained by the MCNP, are shown in figure 2.

Comparison of the results of the analytical calculation with the results of calculations made using the MCNP shows that in the analytical method, the dose rate at a distance up to 1 m from the source is 30% higher than the values obtained by the program. Most likely this is due to the geometry of the calculations - a volumetric source was prescribed according to the program, with all the impurities, that is, taking into account all the processes associated with the scattering and absorption of gamma radiation and electrons in the source and in the ambient air.

According to the radiation safety standards, zoning for three categories of personnel should be carried out from the following considerations:

- The annual dose of exposure for Category A personnel is 20 μSv. If the working hours are set at 1700 hours per year, the dose rate at the workplace of the personnel of this group should not exceed 12 μSv/h.
- the annual dose limit established for Category B personnel (persons, working under effect of technogenic radiation sources according to job conditions) is 5 μSv with an exposure time of 2000 hours per year. Accordingly, the dose rate at the workplace for Ctegory B personnel should not be more than 2.5 μSv/h.
  population and personnel, which is not involved into production activity should not be affected by dose rate more than 1 μSv for 2000-2500 hours per year that corresponds to dose rate of 0.11 μSv/h.

The results of analytical methods were chosen as more conservative when determining safe duration of stay in dose fields for Category A personnel. Table 3 presents safe duration of stay in dose fields for Category A personnel

Table 3. Safe duration of stay in dose fields for Category A personnel.

Distance to the surface of the irradiated sample R(cm)	Safe duration of stay in dose fields (h /year)
1	1.0
5	41.95
100	10228

Analysis of calculation results shows that:

- personnel will be subjected to increased irradiation from the source;
- the main contribution to the dose rate is made by the gamma radiation of the ^{60}Co isotope;
- duration of stay in dose fields should be limited;
- the work should be carried out under the supervision of the radiation monitoring services and the radiation safety department.

3. Methods of management with obtained RW

During the operation of the plant for irradiated beryllium purification liquid, gas and solid RW will be generated. Gas tritium generated in the plant will be converted into chlorhydric acid (liquid RW) as the result of chemical reaction. Liquid waste is considered as radioactive if specific activity of radionuclides more than 10 times exceeds the intervention level values (IL Bq/g), provided in Hygienic standards 18].

Solid RW will include spent radionuclide sources, plant elements, contaminated objects of the external and internal environment, solidified liquid waste, where specific activity of radionuclides

exceeds the values of the minimum significant specific activity (MSSA Bq/g and MSA Bq) provided in Hygienic standards. The specific activity of isotopes is given in table 4.

Table 4. The specific activity of isotopes.

Isotope	MSSA, Bq/g	MSA, Bq
^{3}H	$1 \cdot 10^{6}$	$1 \cdot 10^{9}$
^{60}Co	~1	$1 \cdot 10^{5}$
^{108m}Ag	~1	$1 \cdot 10^{6}$

Figure 3 presents the pattern of radioactive waste generation, which shows all types of wastes generated during operation of the plant for purification.

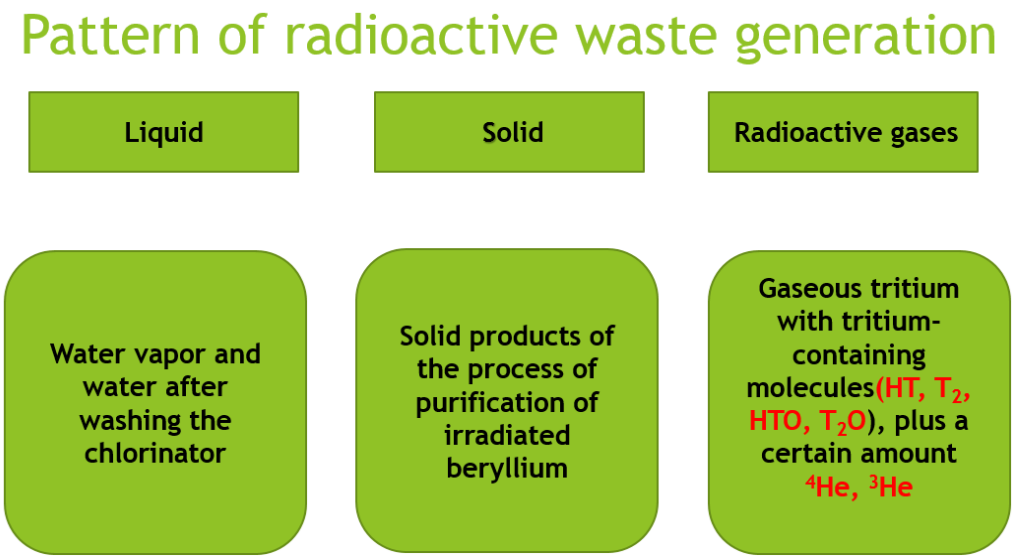

Figure 3. Radioactive waste generation.

Sampling from various parts of the installation were carried out after the experiments. The results of the chemical analysis of products that were in chlorinator after technological purification of irradiated beryllium are presented in table 5.

Table 5. Results of chemical analysis of products of technological process.

Parameters	Sample 1: deposits from the upper wall of the chlorinator	Sample 2: deposits from the lower part of the chlorinator	Sample 3: washing water from chlorinator
Specification:	brown-rusty powder ; the main part is insoluble in water		brownish-brown liquid
Quantity	6.0 g	21.0 g	20 l
Ferrous oxide	mainly consists of oxides of II and III-valence ferrum		insoluble in water
Ferrous chloride and beryllium chloride	5.26 mass %	6.26 mass %	6.39 g/l
Ferrum	0.3 mass %	0.4 mass %	1.65 g/l
Beryllium	0.09 mass %	0.1 mass %	0.3 g/l

The methods to manage the generated RW after purification have been considered. Liquid and solid waste should be divided into three categories according to specific activity:
1) Low active (from 0.001 μSv/h to 0.3 μSv/h)
2) Average activity (from 0.3 μSv/h to 10 μSv/h)
3) High active – more than10 μSv/h

Solid RW is placed into plastic of paper packets which then loaded into collector-containers.

Liquid RW is collected into special vessels. In the case of small amount of liquid RW (less tham 200 l/day) they send for storage of reprocessing. If amount of generated liquid RW is significant (more than 200 l/day), the project is allowed to discharge the waste into special sewer system.

Gas tritium will be converted into chlorhydric acid as the result of chemical reaction.

During the operation of the plant for irradiated beryllium purification liquid, gas and solid RW will be generated, which can be classified as average active. Subsequent RW management is regulated by the requirements of hygienic standards.

4. Conclusion

As the result, the following can be concluded:

1. The key parameters of purification methods of current activity of the most active beryllium block as of 1 July 2018 have been determined. The maximum activity is $^{3}H = 1.81 \cdot 10^{10}$, $^{60}Co = 1.01 \cdot 10^{4}$, $^{108m}Ag = 1.11 \cdot 10^{3}$ Bq;
2. The power of gamma radiation doses from the most active beryllium block at workplaces close to the olant at a distance of 1 m is 3.32 μSv/h, and the activity under these conditions belongs to the third class of works. The personnel involved in the research on the development of the method for cleaning irradiated beryllium will belong to Category A.
3. Calculated duration of works in the area of the plant for Category A personnel at a distance of 1 m can be up to 10228 h/year.
4. During the operation of the plant for irradiated beryllium purification liquid, gas and solid RW will be generated, which can be classified as average active. Subsequent RW management is regulated by the requirements of hygienic standards.

Acknowledgements

The work was implemented under financial support of Public Institution "Science Committee of the Ministry of Education and Science of the Republic of Kazakhstan" under Contract No.305 of 30.03.2018 on the topic "Advancing the dry purification method of irradiated beryllium".

References

[1] Behrens V 2003 *Group VIII Advanced Materials and Technologies: Powder Metallurgy Data* (Berlin: Springer) p 1
[2] Hausner H 1965 *Beryllium its Metallurgy and Properties* (University of California Press) p 239
[3] Tomberlin T 2004 *Beryllium – A Unique Material in Nuclear Applications* (Idaho National Laboratory. Idaho National Engineering and Environmental Laboratory)
[4] Shchurovskaya M et al 2018 *Validation of the MCU-PTR computational model of beryllium poisoning using selected experiments at the IRT-T research reactor* (Annals of Nuclear Energy vol 113) p 436
[5] Petti D et al 2006 *Fusion Engineering and Design* **81** 8
[6] Kolbasov B, Khripunov V and Biryukov A 2016 *Fusion Engineering and Design* **109-111** 480
[7] Omar H et al 2012 *Applied Radiation and Isotopes* **70** 988
[8] Neha Mehta, Giovanna Antonella, Dino, Franco Ajmone-Marsan, Manuela Lasagna, ChiaraRomè, Domenico Antonio De Luca 2018 **622** 900
[9] Mehta N et al 2002 *Waste Management* **22** 549
[10] Baklanova Yu et al 2013 *Kurchatov:Vestnik NNC RK* 3 39 [in Russian]
[11] Baklanova Yu et al 2015 *Method for measuring chlorine content in a stream of beryllium*

chloride: innov. pat.: (19) KZ (13) A4 (11) 30009 (applicant and patent holder is RSE NNC RK no 2014/0121.1).
[12] Baklanova Yu et al 2014 *The device for producing beryllium chloride from irradiated beryllium and the method of its operation: innovat. (19) KZ (13) A4 (11) 30017* (applicant and patent holder is RSE NNC RK. No. 2014 / 0122.1)
[13] Oak Ridge National Laboratory 2004 *RSICC computer code collection, MCNP5C. Monte-Carlo N-Particle Transport Code System* (Los Alamos: Los Alamos National Laboratory)
[14] Radiation Safety Standards 1999 *NRB-99. SP 2.6.1.758-99* (Moscow: Atomozdat)
[15] Golubev B 1990 *Dosimetry and protection from ionizing irradiation* (Moscow: Atomozdat)
[16] Moiseyev A and Ivanov V 1990 *Referencebook of dosimetry and radiation hygiene* (Moscow: Atomizdat)
[17] Radiation Protection 1978 *Recommendations of MCRP* (Moscow: Atomozdat)
[18] Hygienic standards 2015 *Sanitary and epidemiological requirements to ensure radiation safety* (Approved by the Order of the Minister of National Economy of the Republic of Kazakhstan)