Applied Radiation and Isotopes 69 (2011) 1621–1624

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Neutron flux measurements at the TRIGA reactor in Vienna for the prediction of the activation of the biological shield

Stefan Merz [a], Mile Djuricic [a,b], Mario Villa [a], Helmuth Böck [a], Georg Steinhauser [a,*]

[a] Vienna University of Technology, Atominstitut, Stadionallee 2, 1020 Vienna, Austria
[b] Nuclear Engineering Seibersdorf, 2444 Seibersdorf, Austria

ARTICLE INFO

Available online 24 May 2011

Keywords:
Neutron flux density devolution
Neutron flux monitor
Beam tube
Concrete
Neutron activation
Neutron diffusion length

ABSTRACT

The activation of the biological shield is an important process for waste management considerations of nuclear facilities. The final activity can be estimated by modeling using the neutron flux density rather than the radiometric approach of activity measurements. Measurement series at the TRIGA reactor Vienna reveal that the flux density next to the biological shield is in the order of 10^9 cm^{-2} s^{-1} at maximum power; but it is strongly influenced by reactor installations. The data allow the estimation of the final waste categorization of the concrete according to the Austrian legislation.

1. Introduction

The TRIGA Mark II reactor of the Atominstitut is the only operating nuclear facility in Austria. After the first criticality in 1962 it has an average usage of 220 days per year since then. The Atominstitut hosts several research groups, which are related to the reactor, such as reactor physics, radiochemistry, neutron physics and low temperature physics. The reactor has a height of 6.55 m, a length of 8.76 m and a width of 6.19 m. The main construction material is heavy concrete (incorporating barite, $BaSO_4$). The cylindrical reactor tank has a height of 6.40 m and a diameter of 1.98 m. Due to the special TRIGA fuel, it is an inherently safe research reactor and can be operated in pulse mode (250 $MW_{th.}$ for 40 ms). Normal operation, however, is performed at 250 $kW_{th.}$, which corresponds to a maximum neutron flux density of 10^{13} cm^{-2} s^{-1} in the central irradiation tube. For further information on the TRIGA reactor Vienna, see Khan et al. (2010) and Khan (2010). For experiments, several installations of the reactor such as four beam tubes, one thermal column and one dry irradiation chamber can be used.

Until 1999, the 10 MW ASTRA reactor has been operated at the Austrian Research Centers Seibersdorf (Nedelik, 2006). In the course of dismantling, the activity of the biological shield has been determined in order to meet the regulatory requirements for radioactive waste according to the Austrian legislation (Österreichisches Bundesgesetzblatt II, 2006). With the knowledge gained from the decommissioning of the ASTRA reactor, we try to predict the activation of the biological shield of the TRIGA Vienna in the present study.

2. Materials and methods

As a matter of fact, the final activity of any reactor's biological shield can only be measured properly in the course of dismantling of the reactor, because samples of the concrete in the hot areas usually cannot be taken during operation. Based on the activity measurements of ASTRA's biological shield, a mathematical model has been developed that allows the prediction of the activity of long-lived radionuclides in any biological shield as long as some important parameters are known.

In this work, we apply the model to the walls of the TRIGA reactor in Vienna (excluding the reactor floor). The model is based on several reactor-specific parameters, namely the chemical composition of the concrete, data on the operation periods of the reactor (activation and cooling periods), the neutron diffusion length in the concrete and lastly the neutron flux density at the interface of the biological shield and the primary coolant.

From a sample taken from an inactive and easily accessible part of TRIGA's heavy concrete shield, the composition of the biological shield was determined in the course of this study by instrumental neutron activation analysis (INAA). In particular, 216 g of concrete were homogenized in an agate mortar to a particle size of approximately 5 μm and irradiated for 7 h together with geological certified reference materials in the dry irradiation tube of the TRIGA. The concentrations of elements, which yield long-lived γ-emitting activation products is given in Table 1.

Some other elements may be regarded as interesting for the scope of this study, however, they could not be determined in INAA. For example, we take into consideration the formation of long-lived activation products such as ^{3}H and ^{239}Pu. The content of natural lithium in the concrete, yields ^{3}H in the nuclear reaction ^{6}Li$(n,\alpha)^3$H. In analogy, ^{239}Pu is produced by neutron capture in

* Corresponding author. Tel.: +43 1 58801 141389; fax: +43 1 58801 14199.
E-mail address: georg.steinhauser@ati.ac.at (G. Steinhauser).

0969-8043/$ - see front matter © 2011 Elsevier Ltd. All rights reserved.
doi:10.1016/j.apradiso.2011.05.019

natural uranium in the nuclear reaction $^{238}U(n,\gamma)^{239}U$ and the following disintegration of ^{239}U to ^{239}Np and ^{239}Pu in two β^- decay steps. The abundance of lithium in concrete is typically assumed to 3 mg kg^{-1} (Djuricic et al., 2007); for uranium, the detection limit in the INAA (1 mg kg^{-1}) is assumed.

The log books of the reactor provide the model with data on the annual operation, namely activation duration as well as cooling time by radioactive decay. The resulting activity of each activation product in the concrete is given by the activation Eq. (1).

$$A = N\Phi\sigma(1-e^{-\lambda t_a})e^{-\lambda t_d} \quad (1)$$

A is the activity (Bq); N is the number of target atoms (−); Φ is the neutron flux density (cm^{-2} s^{-1}); σ is the nuclear cross section (10^{-24} cm^2); λ is the decay constant (s^{-1}); t_a is the irradiation duration (s), t_d is the cooling time (s).

For the model, at first the activation equation is used without considering a certain value for the neutron flux density, which means that the values obtained are only proportional to the "real" activation. In other words, the value for each radionuclide is yet $A\Phi^{-1}$. The radionuclides considered are ^{3}H, ^{60}Co, ^{134}Cs, ^{133}Ba, ^{152}Eu and ^{154}Eu, which will pose the most relevant activities. Plutonium-239 proved to be formed only in negligible concentrations (Merz, 2011).

In the course of establishing a nuclide vector, all proportional activities are set into relation with one reference nuclide. This normalization makes values for the neutron flux density dispensable, because they are canceled down. In this study, we have chosen ^{133}Ba as a reference nuclide. Activities other than of ^{133}Ba are expressed by applying the isotope-specific factor f, which sets the other nuclides in relation to ^{133}Ba, as exemplified in Eq. (2) for ^{60}Co.

$$A_{^{60}Co} = f_{^{60}Co}A_{^{133}Ba} \quad (2)$$

For materials with multiple radionuclides, the criteria of the Allgemeine Strahlenschutzverordnung (Österreichisches Bundesgesetzblatt II, 2006) are shown in Eq. (3). This equation is used to classify materials with respect to their activity under the light of the necessary radiation protection. The G values (the element-specific regulatory limits), which allow the radiation protection-based classification of the material, are given in Table 2 (according to the Austrian legislation).

$$\sum_i \frac{A_i}{G_i} \le 1 \quad (3)$$

For the nuclides considered in this study, Eq. (3) is hence modified as follows:

$$\frac{A_{^{133}Ba}}{G_{^{133}Ba}} + \frac{f_{^{3}H}A_{^{133}Ba}}{G_{^{3}H}} + \frac{f_{^{60}Co}A_{^{133}Ba}}{G_{^{60}Co}} + \frac{f_{^{134}Cs}A_{^{133}Ba}}{G_{^{134}Cs}} + \frac{f_{^{152}Eu}A_{^{133}Ba}}{G_{^{152}Eu}} + \frac{f_{^{154}Eu}A_{^{133}Ba}}{G_{^{154}Eu}} \le 1 \quad (4)$$

This term is solved for the activity of ^{133}Ba. From this activity, a maximum permissible neutron flux in the concrete can be calculated by using the transformed activation equation. This flux value must not be exceeded for the respective legal classification of the material. In case this neutron flux *was* exceeded, the next higher legal classification with respect to activity or specific activity has to be chosen for the material. The neutron flux – topic of this study – can be measured at all times and can be compared directly with the maximum permissible flux for each classification.

Since the neutron flux usually cannot be measured inside the concrete without applying destructive methods to the biological shield of currently operating reactors, the flux was measured in interesting positions instead, e.g. at the interface between primary coolant and concrete. Based on this data, the flux can be calculated for inaccessible positions using the neutron diffusion length in this material. In a previous report, the neutron diffusion length was measured for concrete with the composition of the biological shield (Krejci, 2007). It was found to be approximately 10.8 cm for neutrons of all energies.

Two neutron flux measurement series at different positions at the TRIGA Vienna are discussed herein (see Fig. 1). The first measurement series was performed at the tank wall between the dry irradiation chamber and beam tube A (sample codes r1.1–r1.5), a second series on top of beam tube B (P1–P6). The beam tubes are made of aluminum (inside the pool) and stainless steel (inside the biological shield) and filled with air. The measurements were done with gold foils as neutron flux monitors (approximately 300 mg, diameter 20 mm, thickness 0.05 mm). For the first series, six foils were mounted on an aluminum rod every 10 cm. The series consisted of five rods. The rods were

Table 1
Relevant chemical composition of the biological shield (concrete) of the TRIGA reactor Vienna measured by INAA.

Element	Concentration (mg kg^{-1})	Element	Concentration (mg kg^{-1})
Co	1.5	Ba	433 000
Cs	2.6	Eu	0.2

Analytical uncertainty < 10%.

Table 2
Excerpt of Anlage 1, Tabelle 1, Allgemeine Strahlenschutzverordnung 2006 (Österreichisches Bundesgesetzblatt II, 2006).

1 **Radionuclide** (Radionuklid)	**2** **Exemption limit** (Freigrenze)	**3**	**6** **Unlimited clearance of** (Uneingeschränkte Freigabe von)	**7**	**10** **Limited clearance of** (Eingeschränkte Freigabe von)	**13** **Half-life in years** (Halbwertszeit in Jahren)
	Aktivity in Bq (Aktivität in Bq)	**Specific activity in Bq/g** (Spezifische Aktivität in Bq/g)	**Solids, liquids with the exception of column 7 in Bq/g** (festen Stoffen, Flüssigkeiten mit Ausn. von Sp. 7 in Bq/g)	**Demolition waste, excavated soil of more than 1000 t/a in Bq/g** (Bauschutt, Bodenaushub von mehr als 1000 t/a in Bq/g)	**Solids for disposal with the exception of column 7 in Bq/g** (festen Stoffen zur Entsorgung mit Ausn. von Sp. 7 in Bq/g)	
Co-60	1E+5	1E+1	1E−1	9E−2	4	5.3
Cs-134	1E+4	1E+1	2E−1	1E−1	6	2.1
Ba-133	1E+6	1E+2	1		30	10.5
Eu-152	1E+6	1E+1	2E−1	2E−1	8	13.3
Eu-154	1E+6	1E+1	2E−1	2E−1	7	8.8

Notation of the columns in the first row is according to this law.