



DETERMINATION OF THE RADIOACTIVITY LEVEL OF CONCRETE USED AS SHIELDING FOR MEDICAL ^{60}Co SOURCE

Irma Bërdufi^{1*}, Erjon Spahiu², Manjola Shytì¹, Elida Bylyku¹

¹Institute of Applied Nuclear Physics, University of Tirana, Tirana, Albania

²Department of Physics, Faculty of Natural Sciences, University of Tirana, Tirana, Albania

Abstract. This study examines the natural and artificial radioactivity in concrete used as shielding material for medical ^{60}Co source temporary stored in our waste storage site. The determination of the radioactivity level is done to see if any leakage or contamination occurred in concrete material after the dislocation of ^{60}Co source to another destination. Concrete samples were taken from the three drums located in the temporary waste storage site and after preparation of samples were placed in a marinelli beaker with a volume of 500 ml and left in isolation for one month to achieve the secular equilibrium. The activity concentrations of ^{40}K , ^{226}Ra and ^{232}Th in ten samples are determined by using gamma-ray spectrometry method with HPGe detector. The average values of activity concentration are found to be $147.56 \pm 6.97 \text{ Bq kg}^{-1}$ for ^{40}K , $18.09 \pm 0.64 \text{ Bq kg}^{-1}$ for ^{226}Ra and $16.90 \pm 0.68 \text{ Bq kg}^{-1}$ for ^{232}Th , respectively. The activity concentration index (ACI) is used as a screening tool to assess the radiological hazard due to possible release of the concrete in environment or to reuse it as building materials. From all analysis performed the maximum value of ACI was 0.21. This value was found to be lower than 1 and in none of them was found the presence of ^{60}Co radionuclide. We conclude depending on the Decision No. 638, dated on 07.09.2016 on the approval of the regulation "On the safe management of radioactive waste in the Republic of Albania" that the concrete could be discharged freely in environment, or it can be used as building material because do not pose any significant risk to humans.

Keywords: radioactivity, waste storage site, gamma-ray spectrometry, activity concentration index, radiological hazard

1. INTRODUCTION

Member states shall ensure the control of building material emitting gamma radiation is within the scope of the European Council Directive 2013/59/EURATOM [1]. The reference level applying to indoor external exposure to gamma radiation emitted by building materials, in addition to outdoor external exposure, shall be 1 mSv y^{-1} according to it. A screening tool is proposed by the directive, for conservatively controlling the radiological hazards due to building materials, using the activity concentration index (ACI) following the equation:

$$\text{ACI} = \frac{C_{\text{Ra}-226}}{300 \text{ Bq/kg}} + \frac{C_{\text{Th}-232}}{200 \text{ Bq/kg}} + \frac{C_{\text{K}-40}}{3000 \text{ Bq/kg}} \leq 1$$

where: $C_{226}\text{Ra}$, $C_{232}\text{Th}$ and $C_{40}\text{K}$ are the activity concentrations in Bq kg^{-1} for radium (equivalent to uranium under secular equilibrium conditions), thorium and potassium, respectively.

The directive says clear that the attention must be done to the cement since the cement is a constituent (construction) material and the final decision must be applied to building materials like concrete, mortar or plaster. Directly applying the Council Directive 2013/59/EURATOM, represent the worst scenario (maximum contribution of cement on the calculation of

ACI for concrete, mortar or plaster) when cement is considered as building material and not as constituent, i.e., considering a mixing fraction of 1. The EU Directive for the purpose of this study is adopted.

The Decision No. 957, date 25.11.2015 "On reference levels of indoor radon concentration and other radio nuclides concentrations in commodities with public protection effect", on the Part IV, article 16 gives the gamma radiation reference levels emitted by building materials [2] and where in the Annex 5 is given the activity concentration index for the gamma radiation emitted by building materials where the activity concentration levels need to be calculated by the formula given. If the ACI is higher than 1, than the public dose should be calculated taking into consideration other factors such as density, thickness of the material type of building and the intended use of the material.

The work in this study is conducted to determine the natural and artificial radioactivity in concrete used as a shielding material for medical ^{60}Co source temporarily stored in our waste storage site. This radioactive source was used in Radiotherapy Department of the University Hospital Center "Mother Theresa" for tumor treatments and after its use was sent to our waste storage site for temporary storage and was shield with a concrete material. After its dislocation to another destination on July 2019 the concrete shield removed from the source was controlled for any contamination with Contamat

*irmaberdufi@gmail.com

FHT 111M equipment. Even no contamination was found, it was decided, to be on the safe side, before released into the environment to collect it inside 200L drums and stored in the waste storage site until the gamma spectrometry analysis is performed. The determination of the radioactivity level is done to see if any leakage or contamination occurred in concrete material after the dislocation of ^{60}Co source to another destination. The main contributors of natural radioactivity in concrete are ^{226}Ra , ^{232}Th and ^{40}K and the artificial radionuclide could also be present is ^{60}Co , resulting from any leakage or contamination.

The aim of this study has been the determination of the activity concentration index (ACI) used as a screening tool to assess the radiological hazard due to possible release of the concrete in environment or to reuse it as a building material and on Decision No. 638, dated on 07.09.2016 on the approval of the regulation “On the safe management of radioactive waste in the Republic of Albania” which gives the release criteria of the radioactive waste materials so that the radiation risks coming from the released material are low enough, so as not to require regulatory control [3].

2. MATERIALS AND METHODS

2.1. Sample collection and preparation

The concrete samples were collected randomly (10 samples) during the end of May 2020 from three drums located in the temporary waste storage site (Table 1) and for each drum is taken three and four concrete samples. The concrete samples were homogenized to a grain size of less than 1 mm and after preparation in a powder form, they were then transferred into a Marinelli beaker with a volume of 500 ml and sealed hermetically. They were weighed accurately and left in isolation for one month to achieve the secular equilibrium.

Table 1. Concrete samples taken from three drums located in the temporary waste storage site in the Institute of Applied Nuclear Physics

No	Sample ID	Matrix
1	12A	Concrete
2	12B	Concrete
3	12C	Concrete
4	12D	Concrete
5	13A	Concrete
6	13B	Concrete
7	13C	Concrete
8	14A	Concrete
9	14B	Concrete
10	14C	Concrete

Sealed samples were left undisturbed for at least four weeks prior to being measured by the HPGe gamma spectrometer to establish a radioactive equilibrium in the ^{226}Ra decay chain segment member of uranium chain, ^{232}Th member of thorium chain and ^{40}K of the three naturally occurring potassium isotopes.



Figure 1. Concrete samples left for secular equilibrium

2.2. Gamma-ray spectrometry measurements

Samples were measured using the high-resolution gamma-ray spectrometry technique, by using a p-type High Purity Germanium (HPGe). Relative efficiency of detector is 40% and energy resolution, Full Width at Half Maximum (FWHM) is about 1.8keV for 1332.5keV from gamma emission of ^{60}Co . Energy calibration was performed using different point sources, whereas the absolute efficiency calibration was performed in the energetic range from 45keV to 2000keV using Laboratory Sourceless Calibration Software (LabSOCS). The absolute efficiency uncertainties vary from 4% at high energies to 10% at low energies [4].

The ^{226}Ra activity concentration was determined by analyzing the two main gamma emissions of radon progenies, ^{214}Pb at 295.2keV and 352keV and ^{214}Bi , at 609.3keV and 1120.3keV, and calculating the weighted average. The activity concentration of ^{232}Th was calculated from 338.4 and 911.2keV of ^{228}Ac and instead, the activity concentration of ^{40}K was determined from its only gamma emission at 1460.8keV. The activity concentration of ^{60}Co is determined by method of gamma-ray spectrometry technique. Two main energetic lines of ^{60}Co of 1332.5keV and 1173.2keV with higher gamma emission were analyzed.

3. RESULTS AND DISCUSSIONS

3.1. Activity concentration

The activity concentrations of natural radio nuclides of ^{40}K , ^{226}Ra and ^{232}Th in ten concrete samples were determined by using gamma-ray spectrometry method with HPGe detector (Table 2).

Table 2. Activity concentrations of natural radionuclides of ^{40}K , ^{226}Ra and ^{232}Th of concrete samples

Sample ID	^{40}K	^{226}Ra	^{232}Th
12A	146.03±6.87	18.65±0.66	16.81±0.67
12B	140.55±6.59	16.85±0.61	15.09±0.62
12C	147.28±6.95	19.78±0.70	18.6±0.74
12D	158.5±7.93	18.28±0.66	16.12±0.66
13A	135.97±6.52	18.04±0.65	16.78±0.69
13B	153.67±7.21	17.16±0.62	19.05±0.74
13C	148.16±6.88	17.66±0.63	16.81±0.66
14A	146.94±6.87	19.74±0.63	17.16±0.68
14B	151.19±7.02	18.39±0.64	15.65±0.63
14C	147.29±6.85	16.43±0.60	16.94±0.67

The range of activity concentrations for ^{40}K is from $135.97 \pm 6.52 \text{ Bq kg}^{-1}$ to $158.50 \pm 7.93 \text{ Bq kg}^{-1}$, the range of activity concentrations for ^{226}Ra is from $16.43 \pm 0.60 \text{ Bq kg}^{-1}$ to $19.78 \pm 0.70 \text{ Bq kg}^{-1}$ and for ^{232}Th is from $15.09 \pm 0.62 \text{ Bq kg}^{-1}$ to $19.05 \pm 0.74 \text{ Bq kg}^{-1}$. The average values of activity concentration are found to be $147.56 \pm 6.97 \text{ Bq kg}^{-1}$ for ^{40}K , $18.09 \pm 0.64 \text{ Bq kg}^{-1}$ for ^{226}Ra and $16.90 \pm 0.68 \text{ Bq kg}^{-1}$ for ^{232}Th , respectively.

These activity concentrations are comparable with the mean concentrations found in Europe, respectively 216 Bq kg^{-1} for ^{40}K , 45 Bq kg^{-1} for ^{226}Ra and 31 Bq kg^{-1} for ^{232}Th [5].

The presence of radionuclide ^{60}Co after analysis of spectrum in two main energetic lines was not found.

3.2. Assessment of radiological hazard

The Activity Concentration Index (ACI) was calculated for all concrete samples using formula 1 below:

$$ACI = \frac{C_{Ra-226}}{300 \text{ Bq/kg}} + \frac{C_{Th-232}}{200 \text{ Bq/kg}} + \frac{C_{K-40}}{3000 \text{ Bq/kg}} \leq 1 \quad (1)$$

where: $C_{226}\text{Ra}$, $C_{232}\text{Th}$ and $C_{40}\text{K}$ are the activity concentrations in Bq kg^{-1} for radium (equivalent to uranium under secular equilibrium conditions), thorium and potassium, respectively.

According to EU BSS (2013), attention must be done since the cement is a composite material and the final decision must be applied to building materials, like concrete, mortar or plaster. Therefore, the ACI values for different cement types express the potential radiological hazard due to use of cement as building hazard, while a mixing fraction factor must be considered to evaluate their contribution [6]. In Table 3 are reported the activity concentration indices (ACI), which varied from 0.18 to 0.21 which are below the screening level of 1, indicating that the annual effective dose criterion of less than 1 mSv $^{-1}$.

Table 3. Activity concentrations Index (ACI) of natural radio nuclides of ^{40}K , ^{226}Ra and ^{232}Th of concrete samples

Sample ID	ACI
12A	0.195
12B	0.178
12C	0.208
12D	0.194
13A	0.189
13B	0.204
13C	0.192
14A	0.201
14B	0.190
14C	0.189

The activity concentration index (ACI) is used as a screening tool to assess the radiological hazard due to possible release of the concrete in environment or to reuse it as building materials. From all analysis performed the maximum value of ACI was 0.21 (Figure 2).

This value was found to be lower than 1 and in none of them was found the presence of ^{60}Co radionuclide. The Decision No. 638, dated 07.09.2016 in the Annex I,

Table II gives the levels of release of solid materials without any further consideration depending on activity concentration of radio nuclides with natural origin. Where, for ^{40}K the activity concentration should be 10 Bq g^{-1} and for every radionuclide from Uranium and Thorium chain the activity concentration 1 Bq g^{-1} . We conclude depending on the Decision No. 638 that the concrete could be discharged freely in environment or it can be used as building material because do not pose any significant risk to humans.



Figure 2. Activity Concentration Index for concrete samples

4. CONCLUSION

The average values of activity concentration are found to be $147.56 \pm 6.97 \text{ Bq kg}^{-1}$ for ^{40}K , $18.09 \pm 0.64 \text{ Bq kg}^{-1}$ for ^{226}Ra and $16.90 \pm 0.68 \text{ Bq kg}^{-1}$ for ^{232}Th , respectively.

The activity concentration of ^{60}Co was analyzed with method of gamma-ray spectrometry technique. In two main energetic lines of ^{60}Co , with higher gamma emission, the presence of this radionuclide was not found. Therefore, there is no contamination in concrete material after dislocation of ^{60}Co source.

The activity concentration index (ACI) is used as a screening tool to assess the radiological hazard due to possible release of the concrete in environment or to reuse it as building materials, from all analysis performed the maximum value of ACI was 0.21.

This value was found to be lower than 1 and in none of them was found the presence of ^{60}Co radionuclide.

We conclude depending on the Decision No. 638, dated 07.09.2016 “On the approval of the regulation for the safe treatment of radioactive waste in the Republic of Albania” that the concrete could be discharged freely in environment, or it can be used as building material because does not pose any significant risk to humans.

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