RADIOLOGICAL CHARACTERIZATION OF THE CONCRETE BIOLOGICAL SHIELD OF THE APSARA REACTOR

by

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The first Indian research reactor, APSARA, was utilized for various R&D programmes from 1956 until its shutdown in 2009. The biological shield of the reactor developed residual activity due to neutron irradiation during the operation of the reactor. Dose rate mapping and in-situ gamma spectrometry of the concrete structures of the reactor pool were carried out. Representative concrete samples collected from various locations were subjected to high-resolution gamma spectrometry analysis. ^{60}Co and ^{152}Eu were found to be the dominant gamma-emitting radionuclides in most of the locations. ^{133}Ba was also found in some of the concrete structures. The separation of ^{3}H from concrete was achieved using an acid digestion method and beta activity measured using liquid scintillation counting. The depth profile of radionuclide specific activity in the concrete wall of the shielding corner was also studied. Specific activities of the radionuclides were found to decrease exponentially with depth inside the concrete walls. This study would be helpful in bulk waste management during the decommissioning of the reactor.

Key words: decommissioning, bulk waste, gamma spectrometry, liquid scintillation, biological shield

INTRODUCTION

APSARA, the first Indian nuclear reactor, a swimming-pool type research reactor employing highly enriched uranium fuel, was utilized for various R&D programmes from 1956 till 2009. Graphite and beryllium oxide encased in aluminum boxes were used as an in-core reflector. The internal dimensions of the reactor pool are: length 8.9 meters, width 2.9 meters, depth 8 meters. Reactor pool water was used as coolant, moderator, reflector, and radiological shielding. The pool walls are made of 2.6 meters thick concrete up to a height of 3 meters from the bottom, tapering to 0.6 meter thick concrete at the top. The reactor was mostly used for radioisotope production, neutron activation analysis, fission studies, neutron radiography, detector testing, seed irradiation, radiobiological research and reactor shielding experiments [1]. The reactor had a maximum power level of 1 MW with a maximum neutron flux of 1 10^{12}/cm^{2}s and was mostly operated up to 400 kW till its shutdown in 2009. In case of the decommissioning of the reactor structure, it is essential to have a detailed radionuclide characterization of concrete in order to assess the type and quantity of specific radioactivity in bulk wastes so as to decide upon the suitable disposal option [2]. Neutron activation of trace element impurities present in the concrete leads to the build-up of activation products, viz. ^{60}Co, ^{152}Eu, ^{154}Eu, ^{133}Ba, ^{3}H, ^{63}Ni, ^{55}Fe, *etc*., in the biological shield. Gamma emitters contribute to a significant external radiation dose in the structural components. ^{3}H ($E_{\beta\max}$ = 18.6 keV) is one of the important hard-to-detect (HTD) radionuclides in concrete which, from the waste management perspective, requires stringent monitoring [3, 4]. The evaluation of scaling factors for difficult-to-measure radionuclides in the structural components of the Indian research reactor CIRUS was carried out previously [3, 5]. Probable modes of production of activation products of concern in concrete are given in tab. 1.

The experiences gained in the decommissioning of the Austrian research reactor, ASTRA, are presented in refs. [6, 7]. Evaluation of residual radioactivity is required for comparison with radionuclide clearance levels [2, 6, 7] for bulk waste disposal. A discussion of regulatory liabilities and environmental remediation in the decommissioning of nuclear reactors in the Czech Republic [8] and publications on the

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Table 1. Important neutron activation reactions considered for concrete

Nuclear reaction	Cross-section [b*]	Daughter	
		Emission	$T_{1/2}$ [year]
$^{59}Co(n, \gamma)^{60}Co$	36	$\beta^- \gamma$	5.3
$^{151}Eu(n, \gamma)^{152}Eu$	9000	EC, X, $\beta^- \gamma$	13.5
$^{153}Eu(n, \gamma)^{154}Eu$	370	$\beta^- \gamma$ X	8.6
$^{3}He(n, p)^{3}H$	5330	β^-	12.3
$^{6}Li(n, \alpha)^{3}H$	940		
$^{2}H(n, \gamma)^{3}H$	0.00052		

*1 b = 10^{-28} m^2

decommissioning plan for the Hungarian research reactor [9] and Romanian research reactor [10] describe the importance of applicable regulatory norms, public acceptance and the possible strategies to be adopted during the actual decommissioning. Detailed radiological dose rate mapping of the APSARA reactor was carried out earlier, during the operation of the reactor [11] and also after shutdown and the dismantling of major reactor components [12]. This work presents the details regarding the methodology and results of the radiological dose mapping and characterization of concrete structures of the pool walls. Depth distribution studies of the residual activity in the walls of the shielding corner side are also presented.

INSTRUMENTS

The instruments employed for characterization studies are described in this section.

Target isotope identifinder GmbH, Germany, (Fieldspec): This is a portable instrument employing a NaI(Tl) detector used for gamma dose rate measurements and radionuclide identification from in-built isotope libraries.

Automess teletector (Model 6150 AD5): This is a commonly used radiation survey instrument employing two GM tubes to span the dose rate measurement range from 0.1 μSv/h to 10 Sv/h. It has a linear response to photons in the energy range of 65 keV to 3 MeV.

Brillance 380 Gamma Spectrometry Detector: Consists of a 38S38 model $LaBr_3$:Ce detector from Saint Gobain Crystals, France, operating at a HV of 551 V, with a resolution of 2.8% at 662 keV. It was coupled to GSpec (Para Electronics, Ltd., India), a portable 14 pin USB-based 1K Wilkinson type MCA.

HPGe gamma spectrometer: This system consists of a coaxial germanium detector, GCD-35 180, from Baltic Scientific. Instruments coupled to an 8K MCA and having a resolution of 1.8 keV at 1332 keV gamma ray of ^{60}Co. PHAST software [13] was used for spectrum analysis.

Liquid Scintillation Counter: The quantification of ^{3}H was done using the Tricarb 2910TR Liquid Scintillation Counter (Perkinelmer Inc. USA) with Optiphase Hisafe-III (Perkinelmer Inc. USA) as the scintillation cocktail.

EXPERIMENTAL

Gamma dose rate mapping and in-situ gamma spectrometry analysis

Gamma dose rate maps of shield cubicles, shielding racks and beam holes were generated using a handheld isotope identiFINDER and automess teletector. Surface dose rates of each square foot of the walls were measured so as to draw the dose rate map of the activated areas. After identifying the hotspots, in-situ gamma spectrometry was carried out by the portable $LaBr_3$:Ce detector at different locations. The portable detector assembly was also used to analyze prominent structural materials like aluminum sheets, MS plates and concrete and lead blocks in the vicinity of the shielding columns.

Sampling and measurement

Representative samples were collected from various locations by drilling the structural components like SS liners, concrete walls of the pool, aluminum liners *etc*. The samples were subjected to gamma spectrometry studies and liquid scintillation analysis to quantify the activity content of long-term activation products of various elements in the structural components.

To estimate the activation products formed in the concrete biological shield, samples of concrete (ρ = = 2.5 g/cm^3) were collected from the shielding corner cavity no: 3 of the APSARA reactor. The sampling location was chosen based on the detailed dose rate mapping and in-situ gamma spectrometry studies. A drilling machine was used to collect representative samples in the form of fine powder. For the radiological characterization of the concrete, sampling was carried out from the surface, at depths of 3", 6", 9", 12", and 18" inside the concrete wall.

Gamma spectrometry measurements

The radioactivity of gamma-emitting radionuclides in the samples was determined using high-resolution gamma spectrometry employing a HPGe gamma-ray detector. Finely powdered concrete samples weighing 25 g were taken in cylindrical vials and gamma radioactivity measured for 10000 seconds. The source-to-detector distance was maintained as 50 mm in all of the measurements. The detector was calibrated using the NPL concrete standard with known activity in the identical counting geometry.

Tritium measurements

^{3}H being a pure beta emitter requires a complete separation from the other interfering β, γ radionuclides

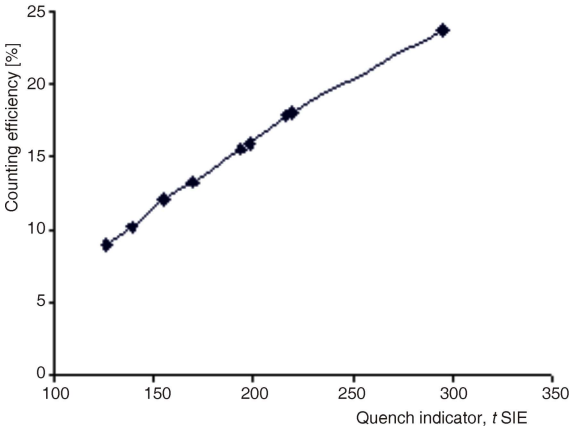

Figure 1. Counting efficiency of ^{3}H for different quench levels (tSIE)

like ^{60}Co, ^{152}Eu, *etc.*, that are present in concrete. Tritium activity was measured using a Tricarb 2910TR liquid scintillation counter. Efficiency calibration of the system was done using the ^{3}H standard. The quench correction curve was constructed in the 0-18.6 keV region using the ^{3}H standard. The quench standard was prepared by adding the required amount of standard to an 8 ml aqueous solution taken up with a 12 ml scintillator in a polyethylene vial. Nitromethane (20 µl to 100 µl) was added as a chemical quencher. Figure 1 shows the counting efficiency of ^{3}H.

The separation of tritium was carried out by an acid digestion method reported earlier [14, 15]. Tritium in the form of condensable HTO collected in the condenser of the digestion system and the gaseous tritium evolved during digestion trapped through bubblers containing acidified water were treated and distilled to facilitate counting with a scintillation cocktail. 12 ml Optiphase hi-safe III scintillator and 8 ml sample was taken for all measurements and counted in LSC for a counting time of 300 minutes.

RESULTS AND DISCUSSION

A typical gamma dose rate map of the APSARA shielding corner side wall after removing the shielding trolley- 3 is shown in fig. 2. The grids with the highest dose rates have been marked in red and those with lowest fields in green. The gamma spectrum obtained during in-situ measurement of the shielding corner wall using the $LaBr_3$:Ce detector assembly is shown in fig. 3. In-situ gamma spectrometry measurements at hotspots of the shielding corner side showed ^{60}Co and ^{152}Eu as the major gamma-emitting radionuclides. It may be seen that the peaks in the in-situ gamma spectrum are not well resolved and hence yielded only semi-quantitative estimates in terms of specific activity ratios. The ratios of specific activities of ^{60}Co with respect to ^{152}Eu present in the structural materials were found to range from 1.1-8.6. It was also noticed that the activity of ^{60}Co was, in general, found to be the highest on MS frames, followed by concrete structures and aluminum and least in lead blocks. High-resolution spectra obtained by HpGe spectrometry were used for accurate quantification and depth profile studies.

Depth profile studies

The concrete samples analyzed for the depth profile study of ^{3}H and other gamma emitters were collected from the shielding corner cavity walls where the dose rate was found to be 40-50 µSv/h. Figure 4 shows the high-resolution gamma spectrum of the neutron-activated concrete sample collected from a 3" depth showing prominent gamma ray peaks of ^{60}Co and ^{152}Eu. The spectrum also shows 873 keV and 1274 keV gamma lines of ^{154}Eu. Though the specific activity of ^{154}Eu was

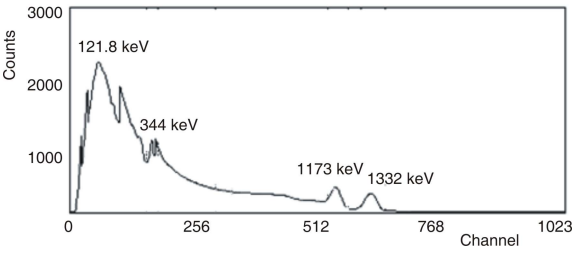

Figure 3. In-situ gamma spectrum acquired using the portable $LaBr_3$: Ce system in the shielding corner wall

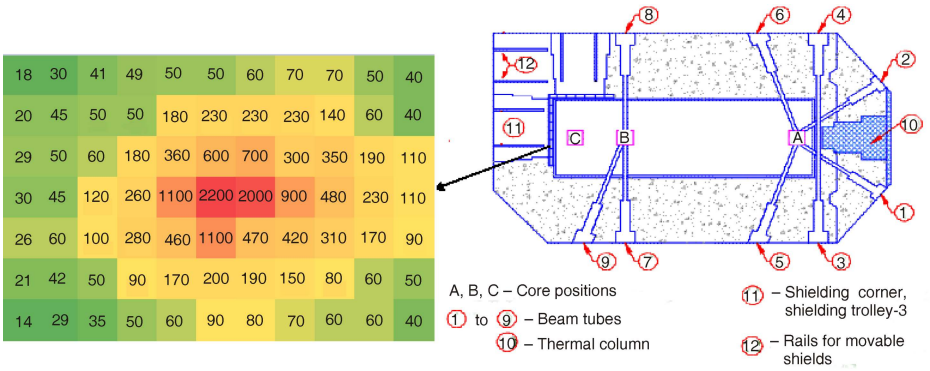

Figure 2. Gamma dose rate map of the wall on the shielding trolley # 3 side [mSvh^{-1}]

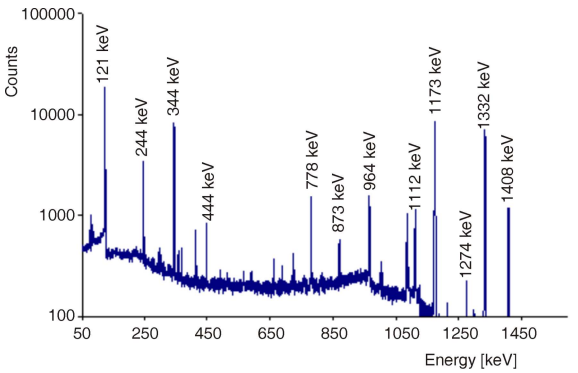

Figure 4. HPGe gamma spectrum of activated concrete bio-shield

negligible, it was observed in the samples up to a depth of 6" inside the wall, after which the levels were too low for detection. Figure 5 shows the LSC spectrum of ^{3}H separated from the concrete sample, along with a background spectrum. Based on the HPGe and LSC analysis of these samples, the depth distribution of the specific activity of the three major radionuclides, ^{3}H, ^{60}Co, and ^{152}Eu, in the shielding corner cavity 3 of the pool is shown in fig. 6. It is seen that the maximum specific activity of ^{60}Co and ^{152}Eu at the surface was found to be 40.8 Bq/g and 33 Bq/g, respectively. Appreciable levels of ^{60}Co and ^{152}Eu were observed up to a depth of 18" inside the concrete blocks near the shielding corner cavity. The specific activity of these radionuclides was observed to follow an exponential function of the depth inside the wall. The exponential distribution of the specific activity with depth in the side concrete wall has also been reported earlier [6]. ^{133}Ba was observed only in surface samples and, hence, not included in the depth distribution graph. The maximum specific activity of ^{133}Ba measured among the surface concrete samples was 20 Bq/g.

Tritium measurement and correlation with γ emitters

A comparison with the background spectrum (fig. 5) shows a complete separation of ^{3}H from other interfering activation products, viz. ^{152}Eu, ^{60}Co *etc.*, present in concrete. The depth profile of the specific activity of β, γ emitters with increasing depth inside the concrete wall was also found to follow an exponential function (fig. 6). This trend is expected, based on the exponential decrease of the neutron flux with depth inside the concrete.

Correlation ratios of ^{3}H activity with respect to ^{60}Co and ^{152}Eu activities observed at the experimental locations are shown in fig. 7. It is seen that the ratio of ^{3}H: ^{60}Co activities ranges from 1.5-3.2 among the samples analyzed. The ratio of ^{3}H with respect to ^{152}Eu specific activities varied from 1.6-4.1. Correlation ratios of DTM radionuclides would help in evaluating

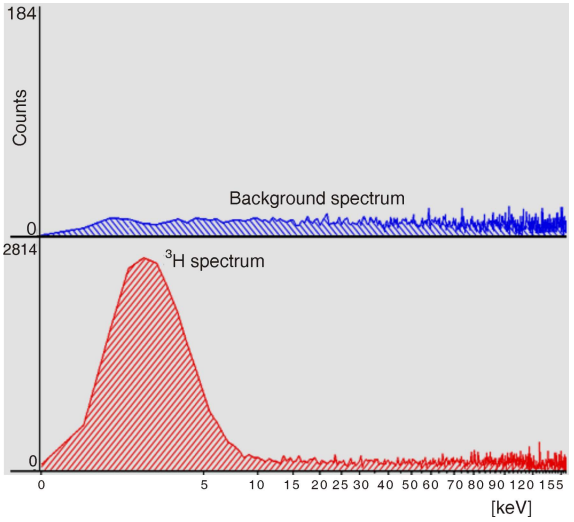

Figure 5. LSC spectrum of ^{3}H separated from concrete

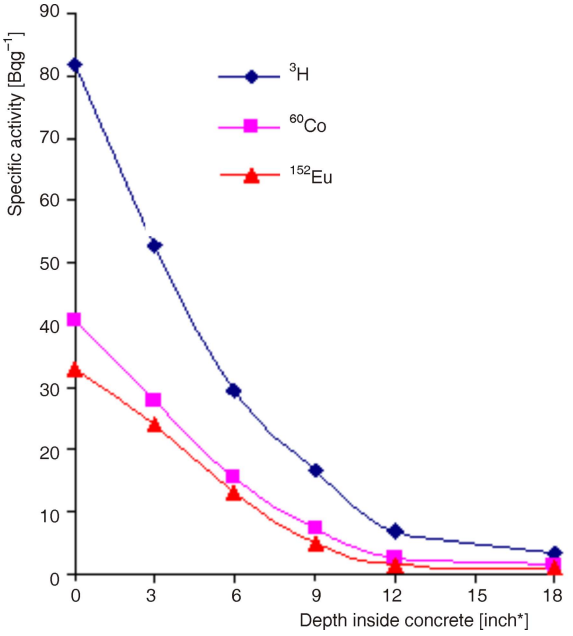

Figure 6. Depth profile of activity inside concrete (*1 inch = 0.0254 m)

the total activity in structural components [2, 16] and in ensuring compliance with bulk waste disposal regulations.

CONCLUSIONS

Based on gamma dose rate mapping and in-situ gamma spectrometry studies, representative concrete samples were collected from different locations of the APSARA pool wall. The samples were analyzed for the quantification of gamma-emitting radionuclides using a high-resolution gamma spectrometry system. Tritium activity in the samples was estimated using liquid scintillation spectrometry. The study of residual

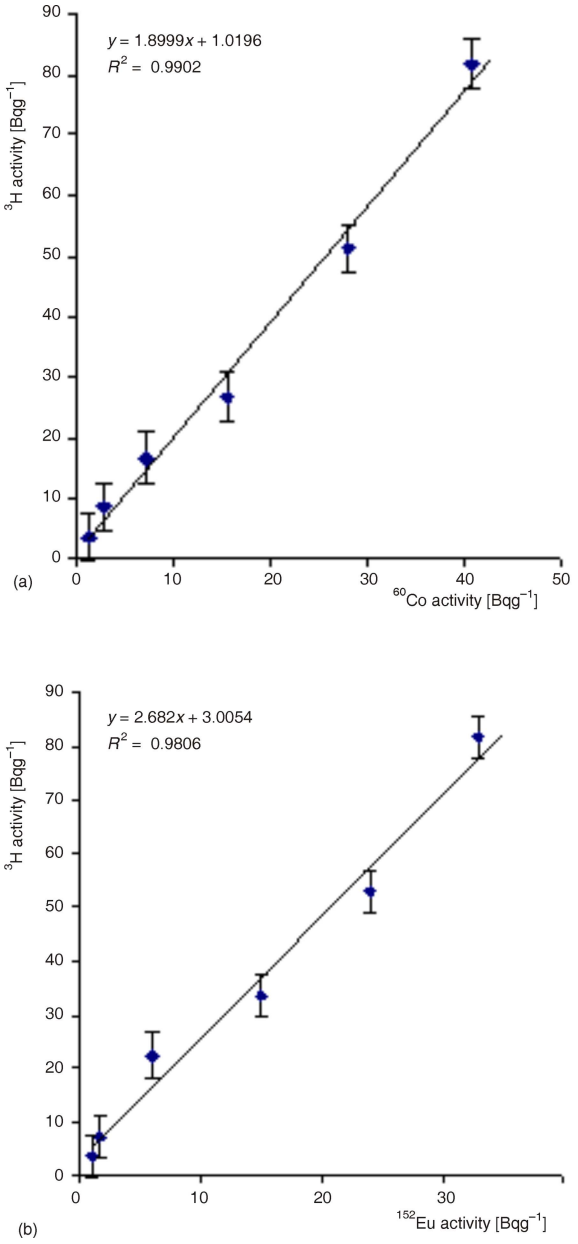

Figure 7. Correlation between specific activities; (a) ^{3}H and ^{60}Co, and (b) ^{3}H and ^{152}Eu

activity of the biological shield of the APSARA reactor showed ^{60}Co and ^{152}Eu as the major gamma emitters that contribute to residual activity. The method adopted for the separation of ^{3}H from other radionuclides present in concrete was satisfactory. Depth distribution of radioactivity in concrete has been carried out as it is useful in active waste volume reduction during the decommissioning phase. This study serves to provide data for developing scaling factors for ^{3}H with respect to the easily measurable correlation partner ^{60}Co and is significant from the standpoint of the clearance of bulk active wastes generated during the decommissioning of the reactor structure.

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AUTHOR CONTRIBUTIONS

The manuscript including tables and figures was prepared by Pr. Srinivasan and Pa. Srinivasan. Experimental analysis of samples using gamma spectrometry system, Liquid Scintillation Spectrometry System, and the depth profile study were carried out by Pr. Srinivasan. Gamma dose rate mapping and in-situ gamma spectrometry studies were conducted by Pa. Srinivasan. Manpower, logistics, co-ordination in dose rate mapping and concrete sample collection were managed by S. Thomas. Guidance in carrying out the dose rate measurements and analysis of experimental results was provided by R. K. Gopalakrishnan. Reviewing and correcting the paper were carried out by A. Goswami.

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РАДИОЛОШКА КАРАКТЕРИЗАЦИЈА БЕТОНСКЕ БИОЛОШКЕ ЗАШТИТЕ РЕАКТОРА АПСАРА

АПСАРА, први истраживачки реактор у Индији, коришћен је у разним истраживачко-развојним програмима од 1956. године до свог гашења 2009. године. Током рада реактора у биолошкој заштити реактора створила се резидуална активност услед неутронског озрачивања. Зато је спроведено мапирање јачине дозе, као и *in situ* гама спектрометрија бетонских структура реакторског базена. Прикупљени су репрезентативни узорци бетона са различитих места који су потом анализирани гама спектрометријом високе резолуције. На већини локација пронађено је да су нуклиди ^{60}Co и ^{152}Eu главни извори гама зрачења. У неким од бетонских структура пронађен је и ^{133}Ba. Сепарација ^{3}H из бетона обављена је методом дигестије киселине, а бета активност мерена је течним сцинтилатором. Разматрана је и расподела по дубини специфичне активности радионуклида у угловима бетонске заштите. Утврђено је да специфична активност радионуклида опада експоненцијално са дубином унутар бетонских зидова. Ово проучавање може помоћи за складиштење отпада великих запремина током декомисије реактора.

Кључне речи: декомисија, отпад велике запремине, гама спектрометрија, течни сцинтилатор, биолошки штит