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Characterization of Radio-Waste from TRIGA II Research Reactor and Assessment of Corresponding Radiation Dose in Bangladesh

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Abstract: During the reactor operation a significant amount of high and intermediate level radioactive wastes are inevitably raised, which are managed and finally disposed of with particular care. The ion exchange resin is used to purify water during the operation of 3 MW TRIGA Mark II research reactor in Bangladesh, when the decontamination efficiency is insufficient to maintain the required primary coolant quality. Five spent ion exchange resin samples and eleven liquid samples from exhausted coolant of 3 MW TRIGA Mark II research reactor were analyzed by using HPGe detector having 40% relative efficiency and 1.8 keV resolution coupled with computer-based MCA. Two radionuclides ^{54}Mn, ^{60}Co have been recognized in the spent ion-exchange resin and three radionuclides such as ^{137}Cs, ^{60}Co, and ^{65}Zn have been identified in the liquid radio wastes samples. The specific activity concentrations of these nuclides and the Inhalation dose for the corresponding nuclide for different body parts such as bladder, Bone surface, Brain, Breast, Stomach, Kidneys, Liver, Red Marrow, lungs, Skin and Thyroid for public are also evaluated according to the ICRP 68. Rad Toolbox software was used for the assessment. The maximum inhalation dose is originated from ^{137}Cs and lung is found to be the maximum radiation sensitive organ, received maximum dose of 53.5 μSv from ^{60}Co (1332 keV). The experiment results would be helpful to develop a policy for radioactive waste management that produced during the operation and maintenance of a research reactor.

Keywords: HPGe Detector, MCA, NAA, Radionuclide, Effective Dose, Ion Exchange Resin, Gamma Spectrometry

1. Introduction

There are 66 Training, Research, and Isotope production by General Atomic (TRIGA) reactors in 24 countries including Bangladesh for diverse applications such as producing radioisotopes for medicine and industry, nondestructive testing, treatment of tumors, for research & education and training [1]. Initially TRIGA was designed to operate with highly enriched uranium but later in 1978 GA launched it as a low- enriched uranium fueling one. Now Uranium-zirconium-hydride (UZrH) is used to operate the TRIGA because of its unique feature like prompt negative temperature coefficient of reactivity, chemical stability and having high probability of retaining fission products [2, 3]. A TRIGA Mark-II research reactor has been installed at the Bangladesh Atomic Energy Commission (BAEC) in 1986 [4]. Since the installment the reactor has been used for radioisotope production (Iodine-131), and various research activities like Neutron Activation Analysis (NAA), Neutron Scattering or Neutron Radiography (NR). The TRIGA Mark-II research reactor at INST, AERE is a light-water-cooled, graphite-reflected reactor operated by ZrH moderator, homogeneously combined with enriched uranium as fuel at a

steady-state thermal power level of 3MW [5]. During the reactor operation a significant amount of high and intermediate level radioactive wastes will inevitably arise, which is managed and finally disposed of with particular care. In TRIGA Mark-II, the ion exchange resins purifier is replaced when the decontamination efficiency is no longer sufficient to maintain the required primary coolant quality. The spent resins are discarded and treated as solid radioactive wastes [6], then is immobilized in cement to avoid escaping radioactivity into the environment. The immobilized wastes are then stored for certain time before discharging to the environment to reach its radioactivity to a levels which are acceptably low [7]. During the processing and storage facilities may cause of escaping radionuclide in nature and environmental contamination which subsequently increase radiation exposure due to inhalation [8-10]. Spent ion exchange materials may require interim storage to allow for radioactive decay if the activity levels in the materials exceed the treatment facility acceptance criteria or if the activity levels in the materials, decay sufficiently in a reasonable time to allow the disposal of the materials without further treatment. Storage for decay should only be used on an interim basis when the radionuclides of concern have a short half-life. Most ion-exchange materials produced in nuclear power plants contain radionuclides with sufficiently long half-lives to preclude this option. For this reason, a study should be conducted on it to ascertain the gravity of the situation and then suggest a mapping to solve it. A lot of works have been carried out in different countries on the spent ion-exchange resin. It is imperative to treat the spent ion-exchange resin and liquid water for the safety of any nuclear research center as well as the human being and the environment. But no study has been conducted for the management of spent ion-exchange resin in Bangladesh though we have a nuclear research reactor TRIGA Mark-II where a huge amount of spent ion-exchange resin and liquid waste produced every year. Therefore, a study is required on spent ion-exchange resin and liquid waste which will influence the design of radioactive waste management for a future radioactive waste disposal center and must be measured inhalation doses for radiation protection for the radiation workers. Keeping this in mind, spent ion-exchange resin from TRIGA Mark-II research reactor, kept in storage room to decay for disposal, were analyzed for radionuclide, radioactivity measurement, determination of the required disposal time and to assess the inhalation dose from different radionuclides for the radiation worker.

2. Experimental Method

2.1. Sample Collection and Preparation

The spent ion exchange resins, which are safely stored (Drum D_1, D_2, D_4, D_5, D_7) in central waste processing and storage facility, were collected in 200 ml plastic container, sealed tightly with cap with proper identification number (R_1, R_2, R_4, R_5, R_7). Liquid samples were collected from TRIGA Mark-II research reactor in plastic container with proper identification code W_1, W_2, W_3, W_4, W_5, W_6, W_7, W_8, W_9, W_{10}, W_{11}. The sample weight and surface dose rate (by using survey meter) of the resin and water samples were recorded. After preparation of the sample the container is wrapped with thick vinyl tape around their screw necks and is kept in the laboratory for counting by the HPGe detector.

Figure 1. *Ion-exchange resin.*

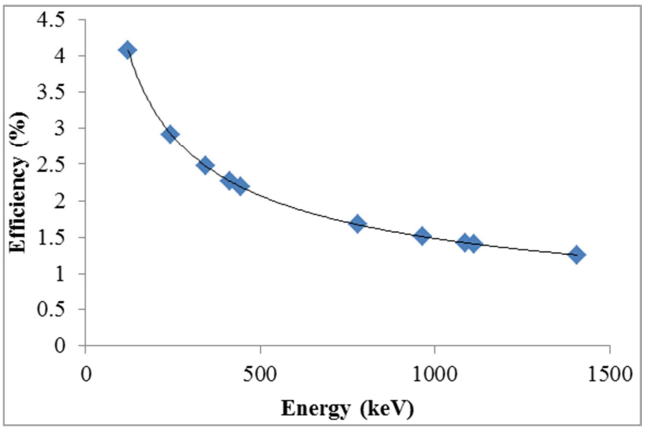

Figure 2. *Energy Calibration curve of the HPGe detector (for resin sample).*

2.2. Gamma Spectrometry

Gamma spectrometry was accomplished by HPGe detector with relative efficiency of 40% and resolution (FWHM) 1.8 keV at 1332 keV γ-energy of ^{60}Co, connected to a MCA.. Although, NaI (TI) detector's efficiency is better than HPGe detector but HPGe is widely used for gamma spectroscopy because of its high resolution [11]. The energy calibration of the HPGe detector [12] was performed by using known energy sources.

$$\varepsilon(\%) = \frac{\text{CPS}}{\text{Activity of the source} \times I_\gamma} \times 100\% \qquad (1)$$

where, I_γ = Absolute gamma ray intensity of the standard source and

CPS = Counts per second = Net peak area per unit time

The formula for the efficiency (%) in terms of gamma energies are therefore,

$$Y = A \times X^{-B} \quad (2)$$

Where, Y, is efficiency expressed in percentage, and X is gamma-ray energy in keV, A, B are constant. The energies response linearly with channel numbers and the MCA was adjusted to suitable channel numbers by entering the energies of the calibration sources in keV into the MCA to convert all channels to respective energies [13]. Gamma spectra detected by the detector was accumulated on MCA and analyzed using Genie-2000 software from Canberra.

2.3. Radioactivity Measurement

Having established the efficiency curve, the measurement of gamma emitting radionuclides in solid spent ion-exchange resin radioactive waste samples were carried out. Considering 835.04 keV, 1173 keV, and 1332 keV, gamma energies, the corresponding radionuclides of ^{54}Mn, ^{60}Co (1173), and ^{60}Co (1332) were recognized. Estimating the counts under the gamma energy peaks of interest, the activity was calculated according to the equation [14-16]:

$$\text{Activity, } A = \frac{CPS \times 1000}{\varepsilon(E) I_\gamma W} \text{ (Bq/Kg)} \quad (3)$$

Where A is the activity in the waste (Bq/Kg), CPS is count per second, ε(E) is the detector efficiency at energy E, I_γ is the intensity of the gamma photon, W is the weight (Kg) of the spent ion exchange resin sample. All the samples were counted for 10,000 seconds and activity concentration in Bq/Kg for the spent ion exchange resin sample have been calculated Table 1 respectively. Then the overall radioactivity (due to the multiple-energy radio-nuclides) was calculated by the following equation.

$$A_T = \frac{\sum_n A_n I_n}{\sum_n I_n} \quad (4)$$

Where I_1, I_2 …. I_n signify intensities of the individual gamma photons and A_1, A_2….. A_n signify the activities at the corresponding energies.

2.4. Effective Dose Assessment

The empirical equation to estimate the effective dose to an individual due to inhalation of radionuclide is [17-19]:

$$E_i = 10^{-6} \nu_i . R_i \sum D_{i,m} . k_m \int_T C_m(t) . dt \quad (5)$$

Where, ν_i = age-dependent breathing rate (m^3/s); R_i is the inhalation reduction factor for staying indoors (dimensionless); $D_{i,m}$ is the dose conversion coefficient (nSv/Bq), k_m the factor accounting for particle size and absorption type for radionuclide, $C_m(t)$ is the time-dependent activity concentration of radionuclide, *i* indicate the specific age group and m indicate the type of radionuclides.

3. Results and Discussions

Using equation (3) the activity concentration in resin and liquid samples have been calculated and presented in table 1 and table 2 respectively. The absorbed doses due to inhalation of each radionuclide, identified in the samples, have been calculated using Rad Toolbox software. The average inhalation doses are tabulated in Table 3. In the resin samples the activity concentration of ^{54}Mn was found to have the range from 34.33 Bq/Kg to 932.78 Bq/Kg with an average value of 341.41Bq/Kg. The activity concentration of ^{60}Co (1173) was found to have the range from 1336.89 Bq/Kg to 15562.02 Bq/Kg with an average value of 7780.466 Bq/Kg and the activity concentration of ^{60}Co (1332) was found only in R_1 which is 16923.06 Bq/Kg respectively. The total activity concentration for each resin sample due to the contribution of each radio-nuclides such as, ^{54}Mn, ^{60}Co (1173), and ^{60}Co (1332) has been calculated by equation (4). Among 5 resin samples the total activity ranged from 59.79 Bq to 782.61 Bq with the average value of total activity 91.64 Bq. In case of liquid waste, the ^{137}Cs was found only in W3 and the activity concentration was found to be 34871.46 Bq/kg. The activity concentration of ^{65}Zn was found to have the range from 33.27Bq/kg to 6034.82 Bq/kg with an average value of 3336.28 Bq/kg. The ^{60}Co (1173) concentration was found to have the range from 143.54 Bq/kg to 12757.54 Bq/kg with an average value of 4730.49 Bq/kg. The activity concentration of ^{60}Co (1332) was found to have in the sample W2 only and value was 10131.12 Bq/kg.

For spent ion exchange resin in the drums D_1, D_2, D_4, D_5, D_7 corresponding to the samples R1, R2, R4, R5, R7, the inhalation dose for Bladder, Bone surface, Brain, Breast, Stomach, Kidneys, Liver, Red Marrow, Lungs, Skin, and Thyroid and average effective dose were measured for the given activity of ^{54}Mn, ^{60}Co (1173) and ^{60}Co (1332). The maximum dose distribution in different organs of the body part was calculated for a member of the public (adult) if they inhaled of these artificial radionuclides. The average inhalation doses (μSv) of different organs for each of the radionuclides in both resin and liquid are given in the table 3. In case of resin, for both ^{54}Mn and ^{60}Co (1332 keV), the highest inhalation dose was found for liver 0.043 μSv and 4.430 μSv, and the averge lowest were found 0.0059 μSv and 1.17 μSv. respectively. For ^{60}Co (1173 keV), the average highest value was observed for Lungs (12.630 μSv) and lowest average value was observed for Brain (0.593 μSv).

For the water samples in the drums W_1, W_2, W_3, W_4, W_6,, W_9, the inhalation dose for Bladder, Bone surface, Brain, Breast, Stomach, Kidneys, Liver, Red Marrow, Lungs, Skin, and Thyroid and average effective dose were measured for the given activity of ^{137}Cs, ^{65}Zn, ^{60}Co (1173) and ^{60}Co (1332).

Table 1. *Activity concentration (Bq/Kg) in spent ion-exchange resin samples.*

Sample ID	Weight of the sample (gm)	Surface dose rate (μSv/hr)	Activity concentration (Bq/Kg)			Total Activity (Bq)
			^{54}Mn	^{60}Co (1173)	^{60}Co (1332)	
R_1	43.04	2.20	932.78	14462.74	16923.06	463.67
R_2	42.52	0.25	34.33	3524.62	-	75.67
R_4	60.22	0.23	57.13	4016.06	-	245.3
R_5	44.72	0.21	-	1336.89	-	59.79
R_7	50.29	0.28	-	15562.02	-	782.61

Table 2. *Activity concentration (Bq/kg) in liquid samples.*

Sample ID	Sample weight (gm)	Surface dose rate (μSv/hr)	Activity Concentration (Bq/Kg)				Total Activity (Bq)
			^{137}Cs		^{60}Co (1173)	^{60}Co (1332)	
W1	155.36	4.36		1638.53	12757.54		1638.53
W2	158.44	3.36		1292.24	8164.9	10131.12	1292.24
W3	139.94	0.53	34871.46	2333.72	1246.7		2333.72
W4	124.33	1.68		30.02	241.5		30.02
W5	105.12	0.20		-			-
W6	143.87	0.12		15.43	143.54		15.43
W7	116.02	0.15		-			-
W8	120.52	0.22		-			-
W9	114.84	0.22		568.95	5828.76		568.95
W10	108.71	0.20		-			-
W11	111.19	0.22		-			-

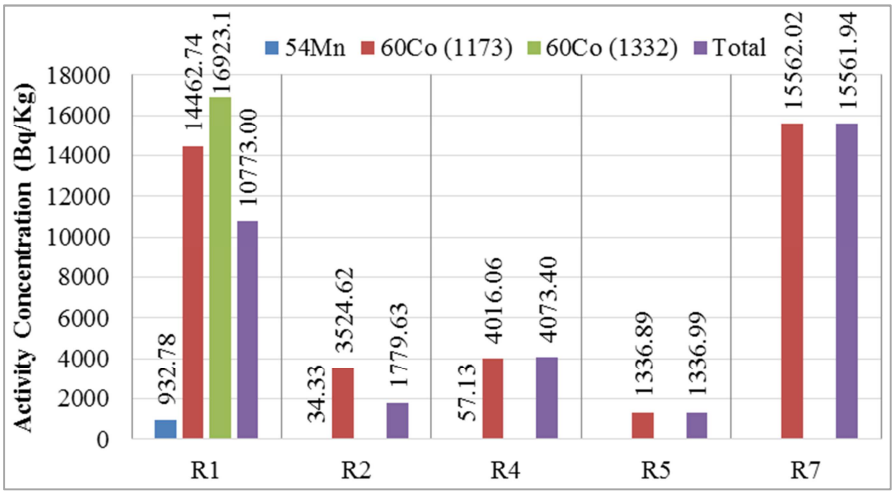

Figure 3. *Average activity conc. in resin sample.*

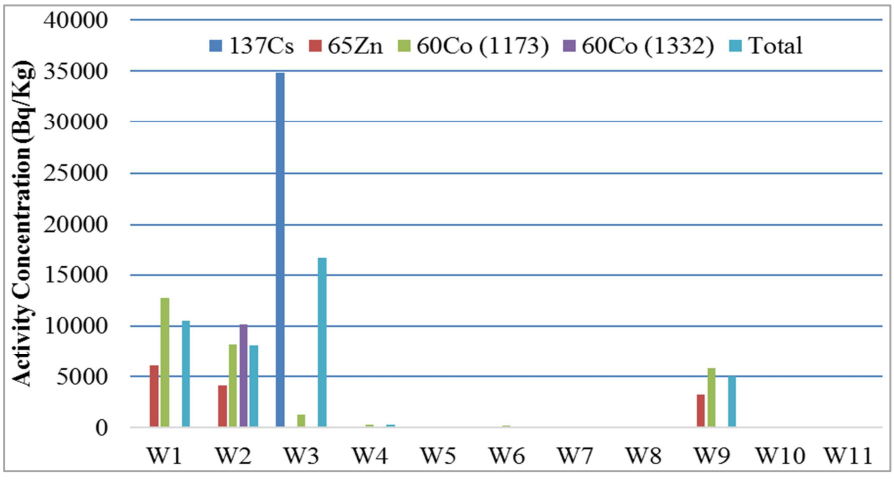

Figure 4. *Average Activity concentration in liquid.*

Table 3. *Average inhalation doses for different organs of the body.*

Body organs	Resin samples			Liquid samples			
	^{54}Mn	^{60}Co (1173 keV)	^{60}Co (1332 keV)	^{137}Cs	^{64}Zn	^{60}Co (1173 keV)	^{60}Co (1332 keV)
Bladder	0.0087	0.853	1.68	33.7	0.782	1.49	3.62
Bone surface	0.0255	1.038	2.04	32.2	1.37	1.77	4.41
Brain	0.007	0.593	1.17	27.8	0.735	1.09	2.52
Breast	0.00647	2.823	2.55	26.4	0.979	2.04	5.51
Stomach	0.0099	1.187	2.33	30.7	0.979	2.06	5.04
Kidneys	0.014	1.115.	2.19	31.7	0.97	2.02	4.72
Liver	0.043	2.265	4.45	31.7	1.127	3.84	9.60
Red Marrow	0.021	1.115	2.19	30.7	1.28	1.95	4.72
Lungs	0.0108	12.630	24.8	29.8	3.18	21.91	53.5
Skin	0.0059	1.231	1.24	25.4	0.636	1.09	2.68
Thyroid	0.0077	1.001	1.97	30.7	0.979	1.74	4.24

The average inhalation doses of different organs for ^{137}Cs, ^{65}Zn, ^{60}Co (1173 keV), and ^{60}Co (1332 keV) in µSv are given in the table 3. For both ^{65}Zn and ^{60}Co (1173 keV) the average highest value was observed for Lungs, which was 3.18 µSv and 21.19 µSv. Similar to resin, for ^{60}Co (1332keV) in liquid, the average highest value was observed for Liver, which was 9.60 µSv and lowest average value was observed for Brain, which was 2.52 µSv. The ^{137}Cs nuclide is found to be the most threat for human being since it provides the highest absorbed dose than any other nuclides observed in both resin and liquid waste.

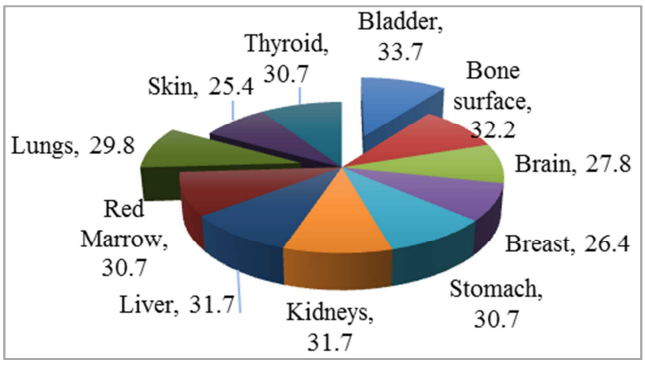

Figure 5. *Inhalation doses (µSv) from 137Cs.*

The average inhalation dose from ^{137}Cs is shown in figure 5. The absorbed dose by lung due to inhalation of different radionuclide is shown in figure 6.

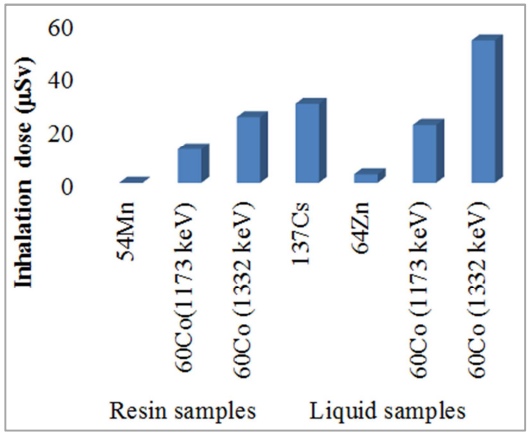

Figure 6. *Inhalation dose (µSv) for Lungs.*

The maximum dose distribution in different organs of the body part was calculated for public (adult) if they inhaled of these artificial radionuclides. From these radionuclides Lungs received high dose from the contribution of these radionuclides. The result of these studies provides the information that could be useful as a baseline data. This baseline data can help to establish national safety culture for radiation protection.

4. Conclusion

Two radionuclides, ^{54}Mn, ^{60}Co have been identified in resin samples and three radionuclides ^{137}Cs, ^{65}Zn, ^{60}Co have been identified in the water sample. The total activity concentration and inhalation doses of radionuclides were calculated to assess the radiological hazards from spent ion exchange resin and water. Among the five identified radionuclide ^{137}Cs provide the maximum inhalation dose and lung found to be the most radiation sensitive organ. In order to establish radiation safety criteria, further study is required to estimate the effective equivalent dose both for public and worker involved in radio waste management.

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