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Estimation of Concentration of Radioactive Elements for the Liquid Waste Pool in Radiochemistry Laboratories in Al Tuwaitha Site Baghdad-Iraq

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Abstract: The aim of this research is estimating the total radioactivity for the High Level Liquid Waste pool (HLLW) in the radiochemistry waste hall in Al-Tuwaitha site. The collected samples from this pool analyzed by using two techniques, the first is gamma spectroscopy system with High Purity Germanium Detector (HPGe) to measure the radioactivity concentration of gamma-ray emitters and to identify their isotopes. The results of measurements show that the average radioactivity concentration of Cesium (^{137}Cs) equal to 2.085 MBq/L. The second technique is gross alpha beta counter by using (LUDLUM 3030) device with zinc sulfide scintillation detector ZnS (Ag) to measure the radioactivity concentrations of the other radionuclides which are not gamma-ray emitters for the same samples that can not be detected in the first method. Cesium-137 has separated from these samples by evaporation process because it also emmit beta particles associated with gamma photons, the remaining activity is (3.0054 MBq/L). According to work history of the radiochemistry laboratories this waste is nuclear fission products for Uranium-235 (^{235}U) isotope, therefore, the remaining activity refers to Strontium-90 (^{90}Sr) radionuclide due to its long half-life.

Key words: Gamma-ray, emmit, laboratories, waste, isotope, half-life

INTRODUCTION

Radiochemistry Laboratories (RCL) was established in 1978 by the SNIA techint-Italy as a part of the Chemical Research Centre in the previous Iraqi Atomic Energy Commission (IAEC) in Al-Tuwaitha site (about 20 km South Baghdad) (IAEA., 1989). The purposes of the Radiochemistry Laboratories were reprocessing of spent fuel that received from IRT-5000 Reactor in laboratory scale (dissolution, separating and purification) for other chemical research and analysis. The spent fuel that used in this facility (RCL) stored from 1973 (Copland and Cochran, 2013). From these processes there are large amounts of radioactive liquid waste with deferent radionuclides as a result from fission products to Uranium-235 (^{235}U) isotope. These wastes were collected in radioactive liquid waste hall which divided in to three pools (High Level Liquid Waste HLLW, Organic Liquid Waste OLW and Low Level Liquid Waste LLLW). Radioactive liquid waste in HLLW pool fill this pool at a level 1.65 m height from bottom, the long and width of this pool are 7, 3.5 m, respectivily. Therefore, the total volume radioactive liquid waste in HLLW pool is 40.425 m^3 that equal to (40425 L). The fission products of ^{235}U isotope are larger than 160 radionuclides with a deferent of half-life, abundance, particles emissions (alpha and beta particles or photons of gamma) and energy. The remained of these radionuclides where taken in to account are (Strontium-90, Cesium-137, Promethium-147, Europium-155 and Krypton-85) as a shown in Table 1 (Cember and Johnson, 2009). The radioactive waste that accumulate in a spent fuel is fission products with the longer lived ones such as ^{137}Cs and ^{90}Sr, being of most long term concern. ^{137}Cs can be detect by gamma spectrometry system because it is gamma-ray emitter while ^{90}Sr can be detected by several techniques such as Liquid Scintillation Counter (LSC) and Cherenckove radiation technique because it is beta ray emitter (Mills, 1995; Taube *et al*., 1975).

Table 1: The main fission products for U-235 with half-lives g>1 year (John *et al*., 2013; Thomas and Rucker, 2009; Cember and Johnson, 2009)

Radionuclide	Half life (years)	Element state	Boiling point (°C)	Activity percentage at time = 0	Activity percentage at time = 45 years
^{85}Kr	10.73	Gas	-153.22	0.027	1.310
^{90}Sr	29.10	Solid	1382	0.206	55.530
^{137}Cs	30.20	Liquid	671	0.155	43.130
^{147}Pm	2.62	Synthetic	3000	0.706	0.007
^{155}Eu	4.71	Solid	1596	0.010	0.015

Gamma activity: The most common aim in gamma ray spectrometry with High Purity Germanium (HPGe)

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detectors is to determine the radioactivity concentration of each gamma emitting radionuclides which is contained in as the activity per unit volume of the sample. The specific activity of individual radionuclides in liquid samples is given by the following equation (Regguigui, 2006, IAEA, 1989):

$$A(Bq.L^{-1}) = \frac{N}{\varepsilon(E_\gamma).I(E_\gamma).t_c V} \quad (1)$$

$$N = N_S - N_B$$

Where:

N = The net peak area count per second (cps unit)
N_S = The net peak area (cps unit)
N_B = The net peak area in the background (cps unit)
$\varepsilon(E_\gamma)$ = The efficiency of the specific nuclide's energy (%)
$I(E_\gamma)$ = The probability of gamma energy (E_γ)
t_c = The counting of the sample in seconds
V = Volume of sample (L)

Gross beta activity: Gross alpha/beta measurement is one of the simplest radiological analysis procedures which is applied usually as a monitoring technique in the field of radiology, environmental and industrial applications. Calculations conduct in order to convert the counts per minute (cpm) into disintegration per minute (dpm) by depending on beta detector efficiency and finally into units of activity per sample such as Bq kg^{-1} and Bq L^{-1}. A blank planchet is used for background count, the dry residue of liquid samples count for gross alpha and beta activities. The subtraction of the background count from sample's count gives the net count of the sample. The activity of gross alpha/beta calculate by using the equation (Biswas *et al*., 2015):

$$dpm = \frac{Net\,(cpm)}{eff} \times 100 \quad (3)$$

Where:

DPM = Alpha/beta disintegration per min
Net (CPM) = Net alpha/beta count per min minus background count
eff = Alpha/beta efficiency percent

The gross beta activity can determine from the following equation (National Exposure Research Laboratory, 1980):

$$A(Bq.L^{-1}) = \frac{B(dpm)\times 1000}{Vx60}$$

Where:

B = Net beta count rate (disintegration per minute)
V = Volume of sample (mL)

MATERIALS AND METHODS

Gamma spectrometry system (Canberra) with 40% relative efficiency as shown in Fig. 1, resolution (>1.8 KeV) based on measurements of 1.332 Mev gamma ray photo peak of cobalt ^{60}Co source was used for measurements to analyze the samples. This system consists of a coaxial High Purity Germanium (HPGe) detector model GC4118, preamplifier, pulse-height analyzer (DSA1000) and Multichannel Analyzer (MCA) with 8192 channel. Both high voltage supply and amplifier device are compact in one unit (DSA1000). The detector shield with a cavity adequate to (10 cm) lead, absorption grid from (1.6 and 0.4 mm) cadmium and copper to reduce radiological background. A library of radionuclides contain the energy of the characteristic gamma emissions of each nuclide were analyzed and their corresponding emissions probabilities is included in the date supplied in the software program (Genie-2000 V.3.2). The activity concentration (Bq/L) of ^{137}Cs was determined from gamma line (661.6 keV) peak (Agbalagba *et al*., 2012; IAEA, 1989).

LUDLUM 3030 device, power voltage AC (95-250) volt was used to measure alpha/ beta activity, It has built in scintillation detector ZnS (Ag) with shield chamber and chrome plated brass, sample tray diameter is 5.1 cm (2 in) as shown in Fig. 2. The counts per minute (cpm) or disintegration per minute (dpm) modes may be enabling to allow the count to be converted automatically in real time to cpm or dpm. The detector is adhered to plastic scintillation material and magnetically shielded

Fig. 1: Gamma spectrometry system

Fig. 2: LUDLUM3030 device

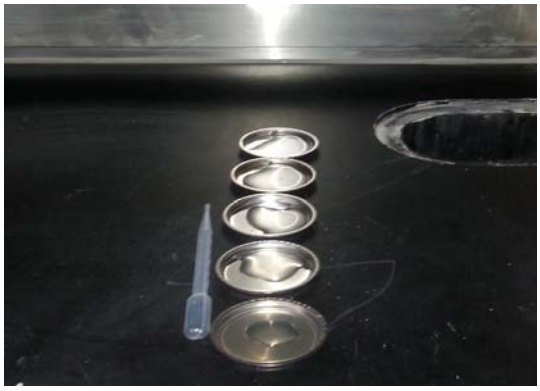

Fig. 3: The stainless steel planchet

photomultiplier, the window 0.4 mg.cm^{-2} type Aluminized Malar, Active and open area (20.3) cm^2, the efficiency (4π geometry) for alpha (0.32-^{230}Th, 0.39-^{238}U, 0.37-^{239}Pu) for beta (0.27%-^{137}Cs and 0.26-^{90}Sr/ ^{90}y) (Kaye and Laby, 1973).

The other materials were used in this research are burn oven with wide range of temperature degree (muffle furnace), hot plate, graduated cylinder (10 mL), beaker (500 mL), filter paper (Whatman 12.5 cm diameter), 1L plastic container, stainless steel planchet (2 in diameter), distilled water and sampling tools.

Samples collection and processing: Three samples (one liter) were taken from bottom, middle and surface from radioactive liquid waste in HLLW pool, samples were coded (HLW-S, HLW-M and HLW-B). Samples transfer to laboratories in plastic containers with secured tape caps. The liquid sampling tools are (electric liquid pump with 0.25 horsepower, flex pipe and 1 L plastic containers). Samples conserved at the time of collection by adding enough 1N HNO_3 to the sample to bring it to pH<2 (15 mL 1N HNO_3 per liter) (National Exposure Research Laboratory, 1980). Liquid samples were filtered by filter paper they put in Marnilli peaker with volume (500 mL) and closed strongly with suitable tape. HLW-M Sample were diluted with distilled water in to five samples, the dilution percentages are 2-10% for gamma spectroscopy system measurements. Five liquid samples from HLW-M-2 sample (4% dilution percentage) with volume (1-5) mL were deposit on a 2 inch stainless steel planchet at 100°C by using a hotplate for gross alpha/beta measurements (Agbalagba *et al*., 2012) as shown in Fig. 3 and 4.

Samples analysis: Gamma spectroscopy system (Canberra) was used to analyze the three liquid samples

Fig. 4: Stirrer hot plate

Table 2: The concentration activity versus dilution by gamma spectroscopy system

Sample codes	Dilution (%)	Activity concentration Bq/L for Cs^{137}	Dead time (%)
HLLW-M-1	2	4.17E+04	3.80
HLLW-M-2	4	8.89E+04	7.87
HLLW-M-3	6	1.38E+05	11.92
HLLW-M-4	8	1.87E+05	15.93
HLLW-M-5	10	2.17E+05	18.28

Table 3: The concentration activity by gamma spectroscopy system

Sample codes	^{137}Cs (Bq/L)	^{40}K (Bq/L)
HLLW-S	1.953E+6	283
HLLW-M	2.085E+6	290
HLLW-B	2.404E+6	301

(HLW-S, HLW-M and HLW-B) as shown in Table 2. HLW-M sample was diluted to five samples (HLLW-M-1, HLLW-M-2, HLLW-M-3, HLLW-M-4, HLLW-M-5) and analyzed as show in Table 3. Gross alpha beta activity by using LUDLUM3030 device was done to determine the activity of strontium-90 (^{90}Sr) by three steps by heating the five deposit samples 1-5 mL, (SA-3) sample was taken in to account with 3mL volume by heating it to 300 and 750°C and measured in each step by gross alpha beta counter and gamma spectroscopy system as show in

Table 4: Gross alpha beta activity by LUDLUM 3030 device at 100°C

Sample codes	Volume	α (dpm*)	β (dpm)
SA-1	1 mL	0	11938
SA-2	2 mL	1	23800
SA-3	3 mL	3	34953
SA-4	4 mL	2	43942
SA-5	5 mL	2	54973

*dpm: disintegration per minute

Table 5: Gross beta activity for SA-3 by LUDLUM 3030 device

Temperature degree (C°)	β (dpm)	β (dps*)
125	34953	582.55
300	35818	596.96
750	21639	360.65

*DPM: Disintegration Per second = 1 Bq

Table 6: The mean of measuring background by LUDLUM 3030 device

Sample code	α (dpm)	β (dpm)	The mean of BG for beta
BG	0-3	100-110	105

Table 7: Gross beta activity dpm to SA-3 sample by LUDLUM 3030 device

Sample code	Temperature (C°)	α (dpm)	β (dpm)	Net beta activity (dpm)-BG
SA-3	750	2	21744	21639

Table 8: Measuring result of SA-3 sample by gamma spectrometry system

Temperature degree (°C)	cps* for Cs^{137} isotope
300	11021
750	739

cps: count per second

Table 4-8. The count for alpha was neglected because it in the background range of LUDLUM 3030 device as show in Table 6.

Cesium-137 and strontium-90 radioactivity: Gamma spectroscopy system was used to measure Cesium-137 activity concentration directly, HLW-M-1 sample with dilution percentage (2%) were depended in measurements, the total activity for ^{137}Cs radionuclide is (2.085 $MBq.L^{-1}$) after integrated to initial concentration (100%) as show in Table 3. LUDLUM 3030 device was used to measure the gross beta activity for (SA-3) sample, it was 21639 (dpm) at 750°C by using Eq. 3, then converted to 120216.66 $Bq.L^{-1}$ by using Eq. 4 as show in Table 4 and 5. The last result represent the total activity to the (SA-3) sample which is diluted by distil water (4%) from the main sample (HLLW-M-2), therefore, the total beta remaining activity is 3.0054 $MBq.L^{-1}$ after integrated to initial concentration (100%). The correction factor for cesium activity is the percentage for SA-3 activity before heating at 750 and after that, it is equal to (6.7%) this factor multiply by the total remaining activity for beta and subtract from it to explain the real activity for strontium-90 radionuclide as shown in Table 8, therefore, the total beta remaining activity is 2.804 $MBq.L^{-1}$ this result is activity for strontium-90 radionuclide (Fig. 5-8).

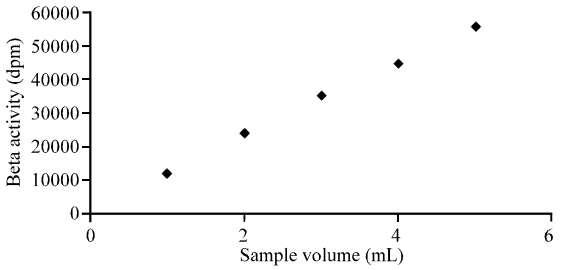

Fig. 5: The deposited liquid sample with beta activity (dpm)

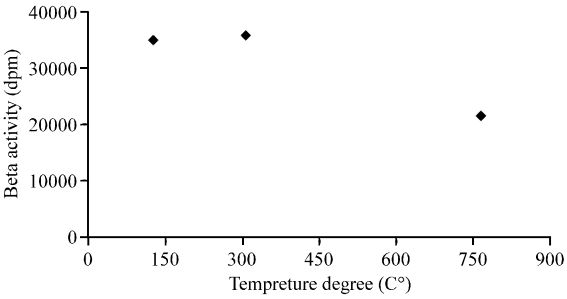

Fig. 6: Gross beta activity dpm for SA-3 sample with temperature degree (°C)

Promethium-147 and europium-155 radioactivity: The total activity for the others radionuclides (^{147}Pm and ^{155}Eu) were calculated theoretically. The percentages of the radionuclides of (^{147}Pm and ^{155}Eu) are (0.007 and 0.015%), respectively.

$^{90}Sr/^{137}Cs$ ratio: Table 1 shows that the theoretical result of the $^{90}Sr/^{137}Cs$ activity ratio is 1.283 from fission products of ^{235}U. In this research it was found that the $^{90}Sr/^{137}Cs$ ratio practically is 1.344 as can be seen in Table 9 and 10, the percent error is 4.75%.

Dissection: The concentration of Cesium-137 isotope can be detected and determined by using gamma spectrometry system but Strontium-90 (^{90}Sr) isotope cannot detect by the same technique because of it is a pure beta emission. Cesium is volatile at elevated temperatures above 450°C (Taube *et al*., 1975). In this research, evaporation of ^{137}Cs from sample was used to separate it from liquid to determine the activity of ^{90}Sr due to their differences in boiling points as show in Table 1. Gross alpha /beta counter (Ludlum 3030) device can be used to determine ^{90}Sr concentration because it has high energy for beta particles. ^{155}Eu isotopes not detected by gamma spectrometry system because of a long time from its product about forty five years that mean nine half life and low concentration in the solution in this time, ^{147}Pm

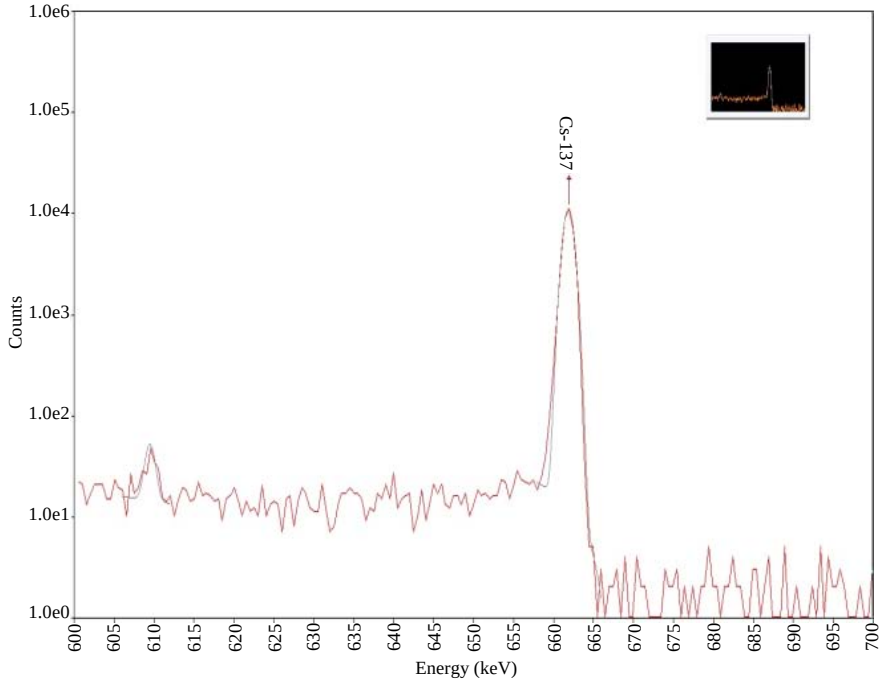

Fig. 7: Spectrum of S-3 sample at 300°C

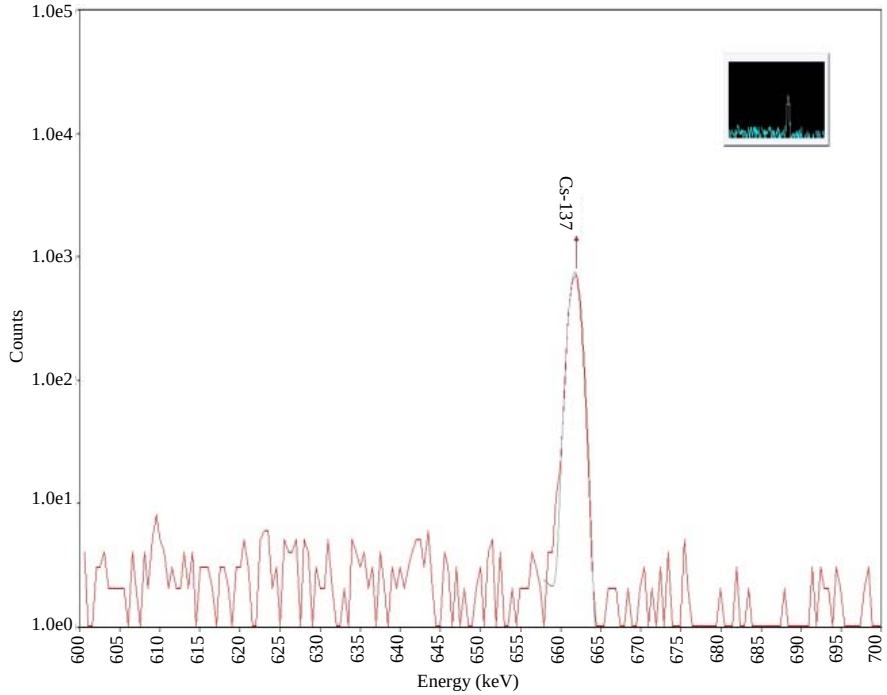

Fig. 8: Spectrum of S-3 at 750°C sample

Table 9: The total activity for high liquid waste HLLW

Volume of HLLW (L)	Activity concentration for ^{137}Cs (MBq.L^{-1})	Total activity for ^{137}Cs (MBq)	Activity concentration for ^{90}Sr (MBq.L^{-1})	Total activity for ^{90}Sr (MBq)	Total activity ^{137}Cs+^{90}Sr (MBq)
40425	2.085	84286.125	2.804	113351.7	197637.825

Table 10: Total activity to all radionuclides in HLLW pool

Activity MBq for ^{137}Cs	Activity MBq for ^{90}Sr	Activity MBq for ^{147}Pm	Activity MBq for ^{155}Eu
84286.13	113351.71	1383.46	2964.56

isotope not detected by the same technique because of it has low gamma probability and low concentration in the solution as show in Table 1. ^{155}Eu and ^{147}Pm are beta particles emitters with low energy, therefore, they can not detected by alpha beta counter (Ludlum 3030) device because of the efficiency to this device very low to these energies therefore, their concentration can calculate theoretically by their abundance in fission products, krypton-85 is not found in this liquid waste because it is gas, so it will be evaporated directly. The gamma spectrometry system results were approximately equal to each other of the selected samples HLW-S, HLW-M and HLW-B. HLW-M sample was depended in measurements because it represents the sample in the middle of pool as well as it represents about the mean activity concentration value between surface and bottom level, it was diluted to five samples with dilution percent (2-10%) in order to overcome the problem of the dead time during the measurement of the samples. The five samples were analyzed using gamma spectroscopy system, the results in Table 3 shows that the dead time was increased from 3.8-18.28% with diluting percentage values of contaminated liquid from 2-10%, respectively the acceptable value of the dead time should be <5%. The first sample (HLLW-M-1) was depended with 2% dilution percent because of it had an acceptable dead time, the activity concentration integrated to ratio 100% as show in Table 3. The results show that the gross beta activity were increased with increasing the deposit volume (Table 4 and Fig. 1). From the physical proprieties of elements (Table 1), the boiling point for Cs^{137} is 671°C, therefore, the sample (SA-3) was heated with 125, 300 and 750°C for 1 h to confirm evaporation of Cs^{137}. SA-3 sample was measured by gamma spectrometry system directly in each step to determine the difference in activity of the ^{137}Cs radionuclide, the results shown there are decrease in ^{137}Cs activity about fifteen times when the sample heated at 750°C as show in Table 8.

CONCLUSION

In the present study, the results indicate that the radioactivity concentration of ^{90}Sr is larger than ^{137}Cs in High Level Liquid Waste pool (HLLW) which is consistent with the theoretical results of fission products yields. The most of ^{137}Cs concentration activity decreased at 300-750°C because the last degree is above boiling point to the cesium element and below the boiling point of strontium elements.

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