



Determination of ^{238}Pu and $^{239+240}\text{Pu}$ in Marine Sediment Using Alpha Spectrometry

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ABSTRACT

A radiochemical separation method to determine plutonium (Pu) radioisotopes in environmental samples has been established in Radiochemistry and Environmental Laboratory in Malaysia. The analysis involving sample digestion with aqua regia solution and followed by radiochemical separation using ion exchange method. The source for alpha spectrometry was prepared by electrodeposition. The chemical recoveries were determined using ^{242}Pu tracer. The method was tested on reference materials and on real sediments from Malaysia marine environment. The validity of the method was checked for coherence using the t-test, relative error and relative standard deviation. All the results obtained were in good agreement with the reference values which indicated that the recommended procedures was successfully applied for the determination of plutonium activities in Radiochemistry and Environmental Laboratory in Malaysia.

Keywords: plutonium radioisotopes, radiochemical separation, alpha spectrometry, sediment

INTRODUCTION

Determination of plutonium (Pu) in environment is become necessity due to the human activities such as nuclear weapon testing, reprocessing nuclear fuel facilities and nuclear power plant accident [1]. The most recent Fukushima Daiichi nuclear power accident had happened in 2011 in Japan due to the Great East Japan Earthquake [2]. This accident had caused in the release of massive radioactive materials into the atmosphere as well as marine environment. By 2014, the research conducted by Fukushima University's Institute of Environmental Radioactivity estimated that the total amount of ^{137}Cs released was 20.5 PBq, 17 PBq to the air and total of 12 to 15 PBq was resolved in the Pacific Ocean [3]. It was estimated that the contaminant to arrive to Straits of Malacca, Malaysia by January 2017 using a modelling Erica software [4]. Therefore, it is very crucial to develop an analytical method to determine Pu radioisotopes in the environment especially in the marine environment.

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Table 1. The half-lives and main alpha energies of respective actinide isotopes [10]

<i>Radionuclides</i>	<i>Half-life, $t_{1/2}$ (Years)</i>	<i>Main Alpha Energy (MeV)</i>
<i>Pu-236</i>	2.90	5.7677
<i>Pu-238</i>	87.76	5.4993
<i>Pu-239</i>	2.41×10^4	5.1562
<i>Pu-240</i>	6.57×10^3	5.1683
<i>Pu-241</i>	14	5.2000
<i>Pu-242</i>	3.74×10^5	4.9005

Furthermore, Malaysia is planning to embark on the Nuclear Power Plant (NPP) by 2030 as reported in the Malaysia Star Online newspaper [5]. In addition, the neighbouring countries to Malaysia such as Vietnam, Thailand and Indonesia are moving forward to adopt NPP as the alternative for power generation. Hence, the determination of Pu in the environment not only beneficial for routine monitoring but also it is very crucial in preparing for emergency purposes.

Several researches have been conducted and reported on the activity concentration of plutonium related to nuclear facilities. For example, 1.2×10^{14} Bq of ^{238}Pu , 2.2×10^{16} Bq of ^{241}Pu and 6.1×10^{14} Bq of $^{239+240}\text{Pu}$ was discharged from the reprocessing plant in Sellafield site [1, 6]. Meanwhile, the total of 1.4×10^{14} Bq of Pu radioisotopes was estimated from the reprocessing plant in La Hague site [7]. In addition, the total of 6×10^{15} of Pu radioisotopes reported from the nuclear power plant accident in Chernobyl in 1986.

Moreover, Pu may exist in different form of isotopes and sources. **Table 1** summarized the half-lives and their main alpha energies respectively. The identification of Pu radioisotopes can be traced back to their sources. The isotope ratios can be used to identify what reactor type and nuclear fuel burn up [1, 8, 9] that had been used for example the study by Bisinger et al. determined the ratio of $^{240}\text{Pu}/^{239}\text{Pu}$ to indicate the presence of plutonium emitted from Chernobyl accident.

Basically, plutonium is formed when the nucleus of uranium atom captures one or more neutrons, changing the atomic structure by creating a new element. In pure form, plutonium is a heavy, silver-coloured radioactive metal about twice as dense as lead [11]. Alpha emitting particles can be very toxic to the human and animal because it can accumulate in the bones and liver. The main pathways of the exposure from alpha emitting particles are from inhalation and ingestion. Most of Pu radioisotopes have very long half-life [12] and therefore can stay longer in animals and humans. Failure to monitor the environment can cause negative impacts in all levels such as economic activities, from domestic to commercial fisheries, fish processing, aquaculture operations as well as tourism. Therefore, it is very important to develop the analytical method of determining plutonium in the environment as it may cause hazard to public and the environment.

The main purpose of this study is to develop a reliable analytical method to determine the activity concentration of plutonium in marine sediment. The ability to isolate Pu

radioisotopes in the marine samples will reflect on the capability of Malaysian Radiochemistry and Environment Laboratory to conduct the radiochemical separation technique with a high level of accuracy.

EXPERIMENTAL

The IAEA reference samples (SRM), IAEA 384 (Fangataufa Lagoon sediment), IAEA 385 (Irish Sea sediment) and IAEA 368 (Pacific ocean sediment), were used to determine the activity concentration of ^{238}Pu and $^{239+240}\text{Pu}$ isotopes in the marine sediments.

Chemicals and Reagents

All reagents used in this analysis were of analytical grade and supplied from Merck brand. The anion exchange resins used were AG 1-X8 (100-200 mesh, chloride form) and AG 1-X2 (50-100 mesh, chloride form) supplied by Eichrom Technologies, Inc. Both resins were preconditioned with 30 mL of 8 M HNO_3 prior to be used. The ^{242}Pu tracer solution with an activity concentration of 98.41 mBq/g (Reference date: 5 February 2002) was obtained from the National Institute of Standards and Technology (NIST). Diluted acids such as 8 M HNO_3 , 10 M HCl , 8 M HCl , 50% (v/v) H_2SO_4 , 1% (v/v) H_2SO_4 and 0.7 M HNO_3 were prepared by diluted with distilled water. Other chemicals such as ammonium hydroxide, ^{242}Pu tracer, hydroquinone solution (0.15 g C_6H_4 in 100 ml 0.7 M HNO_3) and ammonium iodide in 10M hydrochloric acid, $\text{NH}_4\text{I-HCl}$ solution (5.1 g NH_4I dissolved in 100 ml H_2O , the solution was mixed with concentrated HCl in the ratio of 71:29 v/v) were also used [13].

Instrumentation

An Octete plus alpha spectrometry consist of fully integrated control system for eight internal detectors was used. All samples were placed at a distance of 3 mm from the detector. Pressure of 10 Torr was used for vacuum chamber. The counting time was set to 259200 s for each sample. Alpha Vision software was used for evaluation of alpha spectrum. Alpha spectrometry was calibrated for energy and efficiency using a prepared mixed radionuclide standard source containing ^{238}U , ^{234}U , ^{239}Pu and ^{241}Am supplied from Eckert & Ziegler Isotope Products that is traceable to National Institute of Standards and Technology (NIST).

Methodology

Sample preparation

The SRM and sediment samples were ashed in an electric furnace with a gradual heating temperature up to 450°C for at least 8 hours to destroy any organic matter. The bulk sample was homogenized by stirring up using spatula. A sub-sample of 0.5 – 1.0 g was weighed using a calibrated microbalance and 200 μL ^{242}Pu tracer was added into the beaker. The tracer was added at the beginning of the procedure to estimate the radiochemical recovery throughout the separation procedure. Then the sample was digested using aqua regia solution [1] in the ratio of 1:3; 10 mL concentrated nitric acid (65% of HNO_3) and 30 mL concentrated

hydrochloric acid (37% of HCl). The sample was leached several times on a hotplate with concentrated HNO₃ and 1 mL of H₂O₂ until the sediment is completely dissolved. The sample was re-dissolved with 20 mL of 8M HNO₃ before continuing with radiochemical separation.

Radiochemical Separation

The sample solution was loaded into anion exchange column filled with the pre-conditioned Eichrom AG 1-X2 resin. After the last drop of the sample, the column was washed with 30 mL of 8M HNO₃ to elute americium, iron and uranium, meanwhile thorium will be removed after washed by 8M HCl [14]. This washed can be collected if interested with respective radionuclides. Then, Pu was eluted using 40 mL of 0.7M HNO₃ and collected in the new labelled beaker. Further purification was done by eluting the column with freshly prepared hydroquinone solution, C₆H₄ (OH)₂ and collected the wash in the same beaker. Hydroquinone is used to reduce Pu⁺⁴ to Pu⁺³. Then, the solution sample was evaporated until dryness on the hotplate. The sample was re-dissolved with 8M HNO₃ before continuing to purification step.

In the purification step, further separation was done using pre-conditioned Eichrom AG1-X8 resin. The sample solution was loaded to the column and washed using 8M HNO₃ after the last drop of sample. 10M HCl was added to further elute thorium from the resin. Finally, Pu was eluted by using 20-25 mL of freshly prepared ammonium iodide in 10M HCl solution [8, 9]. Sample containing Pu radioisotopes was collected in a new labelled glass beaker and evaporated to dryness on the hotplate. Concentrated HNO₃ was added into the glass beaker containing sample to eliminate iodine from the solution. 1mL of concentrated perchloric acid was added to the sample and evaporated to dryness.

Source Preparation

After sample evaporation, 1 mL of 50% H₂SO₄ was pipetted into the samples and sample was electrodeposited on a stainless steel disc (pH=1.5 to 2.2, constant current of 1.0 A, voltage=10 V for 1-2 hours) and counted by alpha spectrometry. For this measurement, the detection limit calculated according to [15] was 0.2 Bq/kg for Pu radioisotopes. The flowchart of the detail analytical procedure for separation of Pu radioisotopes was illustrated in [Figure 1](#).

Calculation of activity concentration for plutonium

The peak areas of plutonium radioisotopes in the 4750 keV to 5850 keV energy region of the alpha spectrum were determined using Alpha Vision software. The ²³⁸Pu, ²³⁹⁺²⁴⁰Pu and ²⁴²Pu peak areas were corrected by subtraction of the ambient background and reagent blank background.

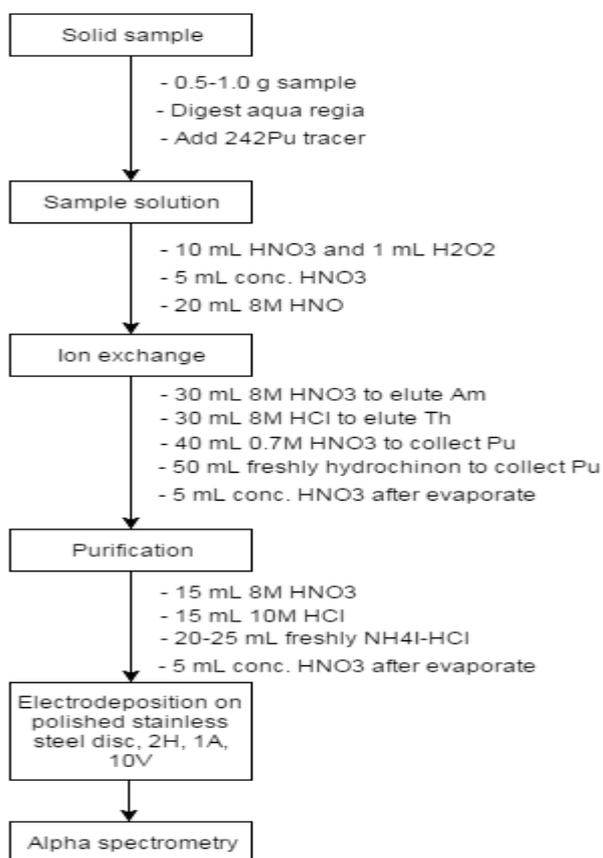


Figure 1. Analytical procedure for ^{238}Pu and $^{239+240}\text{Pu}$ in sediment samples

The activity concentration of plutonium radioisotopes can be calculated using the equation below:

$$A_{\text{Pu}} = \left(\frac{\text{cts} - \text{bkg}}{t} \right) \times \frac{1}{E} \times \frac{1}{\text{Yield}} \times \frac{1}{\text{wt}} \quad (1)$$

where A_{Pu} is the activity concentration of Pu radioisotope of interest, cts is the count of Pu, bkg is the background of the system, t is the counting time, E is the efficiency of the alpha spectrometry system, wt is the weight of the sample and yield is the chemical recovery where it can be calculated using the equation:

$$\text{Yield} (\%) = \frac{\text{net cpm}/E}{\text{dpm added}} \times 100 \quad (2)$$

where cpm is the count per minute and dpm is the activity of the tracer in the disintegration per minute.

Table 2. Values of ^{238}Pu and $^{239+240}\text{Pu}$ (Bqkg^{-1}) in reference materials taken from the International Atomic Energy Agency (IAEA)

Reference material	Certified values ^a		Mean values		RSD (%) ^b		RE (%) ^c	
	^{238}Pu	$^{239+240}\text{Pu}$	^{238}Pu	$^{239+240}\text{Pu}$	^{238}Pu	$^{239+240}\text{Pu}$	^{238}Pu	$^{239+240}\text{Pu}$
IAEA-368	8.5 (7.6-8.9)	31 (29-34)	8.33 ± 0.60 (n = 16)	29.95 ± 2.15 (n = 16)	3.36	4.66	0.17	1.05
IAEA-384	39.0 (38.6-39.6)	107 (103-110)	39.06 ± 1.43 (n = 19)	105.83 ± 3.87 (n = 16)	2.73	4.83	0.04	0.28
IAEA-385	0.44 (0.42-0.48)	2.96 (2.89-3.00)	0.44 ± 0.02 (n = 15)	2.98 ± 0.11 (n = 15)	6.06	1.27	0.07	0.18

n is the number of replicates

^a Certified value and range

^b Relative standard deviation

^c Relative error

RESULTS AND DISCUSSION

The analytical procedure recommended in this study involves sample preparation which includes ashing, digestion, radiochemical separation, source preparation and finally sample counting by the alpha spectrometry. Certified reference materials (SRM) with known concentration and composition of radionuclides are suitable to be used to validate the performance of the test method. A series of experimental was conducted for both SRM in order to verify the procedure used. The leaching procedure with aqua regia solution has been applied by a number of studies for solid environmental sample [12]. Therefore, the acid leaching procedure has been adopted for dissolving the sediment samples.

The elimination of organic materials in the environmental sample is very important in the analysis of Pu because it can cause interference during the radiochemical separation. Thus, it is very important to ash the sample to completely decomposition and destruction of organic materials contained in the sample. In order to determine the chemical recoveries and to minimize unavoidable losses of Pu in the sample, ^{242}Pu tracer was used. Generally, the average chemical recoveries found were 58%, 42% and 29% in IAEA 368, IAEA 384 and IAEA 385 respectively. The lower chemical recovery of Pu in IAEA 385 might be due to very low activity concentration of Pu in the sediment samples.

Table 2 summarized the measured activity concentrations for ^{238}Pu and $^{239+240}\text{Pu}$ in the SRM. The results obtained are in a good agreement when comparing the measured values and the certified values. The energies of ^{238}Pu lie in the range of 5.456 MeV to 5.499 MeV meanwhile for $^{239+240}\text{Pu}$ lie in the range of 5.105 MeV to 5.168 MeV [16]. From the table, it is clear that the results of the analytical method presented were consistent with the reference values. No outlier values are found in any of the activity concentration results for plutonium radioisotopes. These results have shown that the activity values measured in this study are in

a good agreement with reference values and the validity of recommended method is satisfactory.

In IAEA 384, the activity concentration of ^{238}Pu was 39.06 Bq/kg dry wt. which in the ranges of certified value of 38.6 – 39.6 Bq/kg dry wt. meanwhile for $^{239+240}\text{Pu}$ the activity was 105.83 Bq/kg dry wt. also in the certified value ranges 103 – 110 Bq/kg dry wt. The results showed that the mean activity concentration of ^{238}Pu and $^{239+240}\text{Pu}$ in IAEA 385 is 0.44 Bq/kg dry wt. and 2.98 Bq/kg dry wt. respectively whereas in IAEA 368 the activity concentration is 8.33 Bq/kg dry wt. and 29.95 Bq/kg dry wt. respectively.

Furthermore, the precision and accuracy were investigated in terms of relative standard deviation (RSD) and relative error (RE) [14]. The relative standard deviation is derived from the ratio of the standard deviation of the analysis and mean activity concentration of the radioisotopes. The relative error is derived from the difference between the measured values and the certified values of the SRM. A result was only classified as 'in agreement' when the relative error is equal to or not more than 2. There were variations in the RSD and RE for all SRMs might be due to random and systematic error in the radiochemical separation.

Sample t Test

A basic statistic test using 1-sample t test was conducted for the analytical results using Minitab 17 software. The t-test is used to analyse test the results obtained and calculate its confidence intervals in terms of the standard deviation and variance of the results. It is also can determine whether the test results are equal or satisfy with the reference value. The test was conducted for the Pu radioisotopes for IAEA 384 only. For ^{238}Pu , the null hypothesis, μ is 39 which is the recommended value of the certificate or the alternative hypothesis μ is not equal to the reference value.

In addition, the statistical test also conducted using Chi-square and Bonett method. For Chi-square method where the test results were assumed normally distributed, the standard deviation was likely to be between 0.81 – 1.58 Bq/kg and the variance was in between 0.65 – 2.50 Bq/kg. Meanwhile for Bonett method, the standard deviation was in between 0.77 – 1.66 Bq/kg and the variance was 0.59 – 2.77 Bq/kg. The Minitab 17 software used these two methods to calculate a confidence interval for standard deviation and variance. The statistical result was described as below.

Test of $\mu = 39$ vs $\neq 39$							
Variable	N	Mean	StDev	SE Mean	95% CI	T	P
Measured	19	39.062	1.068	0.245	(38.547, 39.577)	0.25	0.803

The test results were analysed with the α -level of 0.05 or 95% confidence interval. From the result, the mean activity concentration for 19 test samples was 39.062 Bq/kg with standard deviation of 1.068. The p-value estimated in this test was 0.803 which means that there is a chance of 80.3% that the data obtained was 39 Bq/kg. In other words, using the recommended

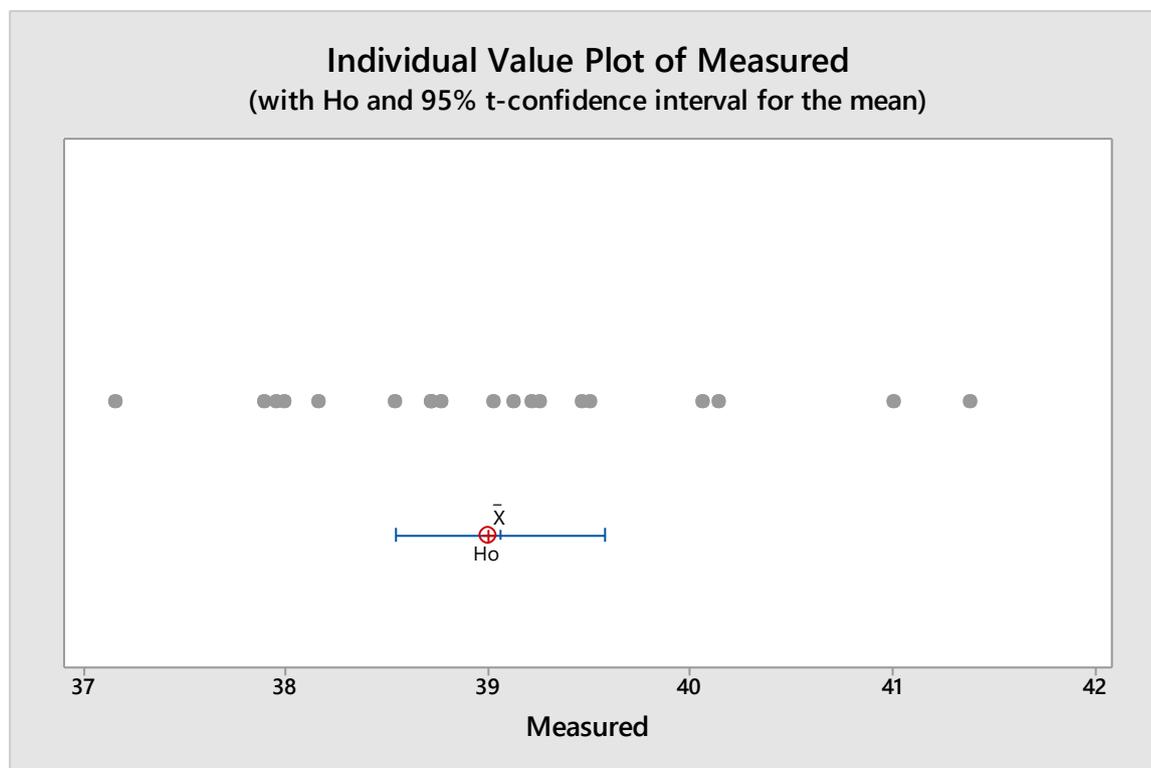


Figure 2. The individual value plot of measured value with null hypothesis and 95% t-confidence interval for the mean

analytical method, the result obtained will be 80.3% of accuracy and in the range of the certificate value.

Figure 2 illustrated the individual value plot of the measured value for ^{238}Pu in IAEA 384. Each dot represented the values obtained from each repetition. The \bar{X} represented the recommended value from the SRM certificate. Therefore, it is statistically proven that using this analytical method, it is likely to obtain precise and accurate result for the marine environment samples.

Marine Samples Monitoring in Malaysia

Malaysia has a total land area of 329,847 square kilometres and a total coastline of 4,675 kilometres. Malaysia coastline borders six major seas, Andaman and Straits of Malacca in the west, Gulf of Thailand and South China Sea in the middle, and Sulu and Sulawesi seas in the east. With its long coast line and the seas surround the country, Malaysia coasts and seas are important contributors to the national economy. Thus, any disaster that occurs at regional seas and coastal zones that have link to the local seas will have some effect on the economy, security and societal well-being of the country.

Several marine expeditions had been conducted in Malaysia in order to evaluate the marine environment in terms of radioactivity elements. The aim of the projects was to expand

and strengthen Malaysian marine radioactivity database to be used as a reference levels in future nuclear activities. The marine environment expeditions has started in 2003 and continuously monitoring until 2013. The obvious example of the usefulness of the database was when the Fukushima accident happened in Japan in 2011. Local researchers and stakeholders were able to assess the impact of Fukushima to Malaysia coastal environment with these databases.

Although the marine environment monitoring for radioactivity has started since 2003, there were still inadequate $^{239+240}\text{Pu}$ inventory within Malaysian waters. In 2003 to 2007, the marine environment monitoring consists of 64 locations including east coast, west coast and Sulu Sea. The results obtained for $^{239+240}\text{Pu}$ in seawater and sediment was consistent with Asia Pacific Marine Radioactivity Database (ASPAMARD) which was 6 mBq m^{-3} and 0.2 Bq kg^{-1} dry weights respectively [23].

The marine environment was continuously monitored for 30 locations within the Exclusive Economic Zone of East Coast Peninsular Malaysia in year 2007 to 2009. According to [18], the concentration of $^{239+240}\text{Pu}$ was slightly higher than the previous study in the southern area. Maybe the reason was due to the 'obstruction' caused by Anambas and Natuna Isles that are located at the southern end of South China Sea which caused the limitation of seawater flow in that area. Seawater flow and eddy pattern might be the factor of the event. However, in the northern zone, the concentration of $^{239+240}\text{Pu}$ was evenly distributed.

The expedition was continued in 2009 to Sabah's Sulu-Sulawesi Sea to collect surface sediment to be analyzed. The average concentration of $^{239+240}\text{Pu}$ in surface sediment was 0.53 Bq kg^{-1} which shows no correlation between the activity and physical parameters [22]. There was no significant different when comparing the result of surface sediment in 2009 with previous study.

The Fukushima Daiichi nuclear power station accident following the Great Tohoku Earth quake that struck Japan in March 2011 has resulted in severe contamination of the marine environment by various radionuclides released from the power station. In short term, sea water contamination was limited to higher latitudes but in long term, this radioactive contamination would be transported and circulated through the wider Pacific Ocean by the ocean currents. As Malaysia coastal zone is linked to Pacific Ocean through South China Sea, Sulu and Sulawesi seas, there was a national concern about the potential impact of these radioactive releases on Malaysian coastal zone and marine environment, which could then have the possible consequences for the coastal communities and the economies of the country.

Another project was started after Fukushima accident in 2011 for 2 years period. Project titled '*The extent and impact of the releases of radioactivity from the Fukushima Daiichi Nuclear Power Plant accident into Malaysian marine environment*' was conducted by Malaysian Nuclear Agency to support an international project by the International Atomic Energy Agency (IAEA). This project was conducted to monitor five permanent stations including Tioman Island, Perhentian Island, Kuraman Island, Bintulu Port and Sandakan.

Table 3. Monitoring data for $^{239+240}\text{Pu}$ for Malaysian marine environment

Year	Location	$^{239+240}\text{Pu}$	
		Surface sediment (Bq kg^{-1})	Seawater (mBq m^{-3})
2003 – 2007 ^a	64 location	< 0.08 - 1.831	NA
	East coast		
	Malacca Straits/West coast		
	Sulu Sea Sabah & Sarawak		
2007 - 2009	30 location	0.21 – 0.45 ^b	< 0.50 - 1.90 ^c
	Peninsular Malaysia Exclusive Economic Zone		
2009	Sabah Sulu-Sulawesi Sea	< 0.10 - 1.65 ^d	NA
	Peninsular Malaysia		
2011 - 2013	Sabah	NA	<1.47 – 4.44
	Sarawak		

NA = Not Available

^a[17], ^b[18], ^c[19], ^d[20]

For each $^{239+240}\text{Pu}$ determination, 200 L of seawater samples were collected at each monitoring station. The sea water was drawn from 1m below the water surface and passed through a water filter ($1\mu\text{m}$) before collected in a 230 L drum container. Pu underwent co-precipitation method using manganese oxide with ^{242}Pu as the recovery tracer in a separate 230 L drum container. Radionuclide precipitates were formed after sufficient stirring and 24 hours settling period. The clear seawater was decanted and slurry precipitates were collected and further treated in the laboratory.

In the laboratory, $^{239+240}\text{Pu}$ slurries were transferred into 2 L beakers for further washing and decantation. The process was repeated for three times to remove the remaining chemicals. The $^{239+240}\text{Pu}$ activity concentration in the collected slurry was determined by the analytical method described in this paper.

Results obtained from the expeditions were tabulated in **Table 3**. There was impossible to compare the sediment result from this study with previous as the data was not available. However, the activity concentration in the seawater was no significant difference between this study and previous data. Thus, marine environment monitoring need to be continued as it is very important and very useful to evaluate the concentration of radioactivity in Malaysian waters.

CONCLUSION

An analytical method to determine activity concentration of Pu radioisotopes was developed for enhancing the analysis capability of Radiochemistry and Environment Laboratory in Malaysia. Three reference materials, IAEA 368, IAEA 384 and IAEA 385 were used as samples and were radio chemically separated. All samples were treated with sample digestion, anion exchange, purification and electrodeposition before being counted using

alpha spectrometry system. The standard reference materials chosen were suitable to determine the activity of Pu radioisotopes to represent the marine environment. The methodology used in the measurement was found to be fairly good to measure the radionuclides of interest. There is no bias for the method to produce an accurate result if activity concentration for Pu radioisotopes. The precision of the method is fairly good with the RSD values which were less than 10%. The average results for SRM were in the range of the certificate provided with the U-score values were less than 2 which indicates that no significant bias of the method. The analytical method also demonstrated that the measuring technique was robust and rugged and presented in statistical point of view in the 1-sample t test. Overall, the analytical technique used can produce a high quality and reliable analytical result for measuring Pu radioisotopes in the marine environmental samples. The activity concentration of $^{239+240}\text{Pu}$ in Malaysia marine environment also presented as they used the recommended analytical method. They were no significant difference of the activity concentration of Pu isotopes in the marine environment between recent study and previous study.

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