ความเหมาะสมของเวลาในการวัดค่า figure of merit และมวลความหนา ของตัวอย่างสำหรับ low-background alpha/beta proportional counter ในน้ำทะเล

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บทคัดย่อ

การตรวจวัดกัมมันตภาพรังสีรวมแอลฟา และบีตาในตัวอย่างสิ่งแวดล้อมที่เป็นของเหลวสามารถดำเนินการได้ด้วยเครื่องวัดรังสีชนิด low-background alpha/beta proportional ชนิดหลายหัววัด (Berthold LB770) นำตัวอย่างน้ำทะเลซึ่งเตรียมด้วยวิธีการตกตะกอนร่วมมาใช้สำหรับหาสภาวะที่เหมาะสมสำหรับการวัด โดยพิจารณาเรื่องระยะเวลาที่เหมาะสม ค่าปริมาณกัมมันตภาพรังสีต่ำสุดที่สามารถวัดได้ (MDA) มวลความหนาของตัวอย่าง (mg/cm-2) และ figure of merit (FOM) ผลการตรวจวัดพบว่า ค่า MDA ที่เหมาะสมปรากฏอยู่ที่ 0.07 Bq/l สำหรับปริมาณรังสีรวมแอลฟา และ 0.05 Bq/l สำหรับปริมาณรังสีรวมบีตา ที่ระยะเวลาการวัด 100 และ 200 นาที ตามลำดับ นอกจากนี้ ยังพบว่าช่วงน้ำหนักของตัวอย่างที่เตรียมด้วยวิธีตกตะกอนร่วมนั้นอยู่ในช่วงที่เหมาะสม ซึ่งสามารถใช้ค่า self-absorption correction factor (F_a) ที่ได้มีการวิจัยแล้ว คือ $F_a = 0.0003w^2-0.0414w+1.692$ เมื่อ w คือ น้ำหนักสุดท้ายของตะกอนในหน่วยมิลลิกรัม

คำสำคัญ : รังสีรวมแอลฟา และบีตา ปริมาณกัมมันตภาพรังสีต่ำสุดที่สามารถวัดได้ (MDA) มวลความหนาของตัวอย่าง figure of merit (FOM)

Optimization of Counting Times, Figure of Merit and Mass Thickness for Low-background Alpha/beta Proportional Counter in Sea Water

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Abstract

The determination of gross alpha and beta activity in environmental aqueous samples can be done by using a low-background alpha/beta proportional counter with multiple detector type (Berthold LB770). The coprecipitate sea water samples were considered for optimal regime of counting time, the minimum detectable activity (MDA), mass thickness of samples (mg/cm^{-2}) and figure of merit (FOM). The result showed that the MDA of the measurements were estimated to be 0.07 Bq/l for gross alpha and 0.05 Bq/l for gross beta at counting time of 100 and 200 minute, respectively. The thickness of samples prepared by coprecipitation technique indicated more parochial range when compared to evaporation to dryness method. In addition, the samples were in the suitable range that can be corrected by the previous self-absorption correction factor (F_a) as $0.0003w^2$-0.0414w+1.692; where F_a is the self-absorption correction factor and w is the weight of the final precipitate in milligrams.

Keywords: gross alpha and beta, minimum detectable activity (MDA), mass thickness, figure of merit (FOM)

1. Introduction

Radionuclides found in the marine environment have mainly two sources of origin which are natural and artificial radionuclides. The naturally occurring radionuclides exist in constant level in a period of time. On the other hand, artificial radionuclides discharged from artificial sources such as nuclear weapon testing, nuclear accident, nuclear reactor, satellite burn-up and waste disposal from nuclear facilities.[1, 2] These have resulted in varying degrees of contamination of the world's seas and oceans.[3] The most well-established chemical separation techniques are widely used for gross alpha determination including the standard method[4] and the coprecipitation technique. Because the amount of the radionuclide present may be very small, carriers are frequently used. The carrier is added in macroscopic quantities and ensures the radioactive species will be part of a kinetics and thermodynamic equilibrium system.

The 10-channel low-level planchet counter LB 770 is one of the detectors that suitable for the quick scanning procedure for environmental survey in monitoring system. To achieve the best sensitivity, the sampling method and counting regimes must be optimized. The results of the optimization are suitable method for sample preparation and the durations of the mentioned time intervals for measurement. Then statistical studies of the preparation and detection process are necessary to observe on design the environmental monitoring systems around nuclear or radiological installations that emit radioactivity into the environment. The minimum detectable activity (MDA), mass thickness of samples (mg/cm^{-2}) and figure of merit (FOM) are used to demonstrate the optimization parameters for low background alpha/beta proportional counter in sea water samples. Important quantities needed for an estimate of FOM and MDA are the counting efficiency (*E*) and the background count rate (B).

2 Materials and Methods

2.1 Sample Collection

Seawater samples were collected from total of 10 sampling sites at Chon Buri province, the Upper Gulf of Thailand, during wet and dry season. The sampling sites are approximately 1 kilometer from coastal area. Five liters of seawater was collected, at 1 meter below the surface water, using a water sampler and stored in a polyethylene bottle. 10 ml of conc. HCl was immediately added[5]. Temperature, pH, salinity, and location of the sampling site were recorded during sampling.

2.2 Sample Preparation

2.2.1 Preparation of Gross Alpha Sample

500 ml of seawater sample was filled in 1000 ml beaker and neutralized by adding 6M NH_4OH. 20 ml of 1M H_2SO_4 was added and boiled, simultaneously stirred with magnetic bar for 3 minutes to eliminate the Rn and CO_2 [6]. The sample was cooled down to room temperature and allowed to stand for 60 minutes before heated to 50 °C. 1 ml of 5 mg/ml Ba^{2+}. Carrier was added to the sample and stirred to precipitate as barium-radium sulphate for 30 minutes at 50 °C in order to obtain optimum precipitate formation. When the solution was at room temperature, 0.2 ml of 0.1% bromocresol purple indicator and 1 ml of 5 mg/ml Fe^{3+} carriers were immediately added to coprecipitate the actinides. Then 6M NH_4OH was added dropwise until the solution's colour changed from yellow to purple. The sample was continued heated and stirred at $50^{o}C$ for 30 minutes and left overnight. The precipitate was filtrated with 47 mm diameter glass filter paper (GF/A, Whatman) using the vacuum filtration system. Finally, the filter was placed on the planchet and dried under infrared lamp. The planchet was kept in desiccator at least 24 hours before being weighed. Radioactivity was measured by using the low background alpha beta counter equipped with multiple detectors. (Berthold LB770).

2.2.2 Preparation of Gross Beta Sample

2,000 ml of seawater sample was filled in 3000 ml beaker. 1 ml of 10 mg/ml Ba^{2+} carrier 1 ml of 10 mg/ml Fe^{3+} carrier and 4 to 5 gram of ammonium chloride were added and heated at 60-70 ºC. The seawater sample was neutralized by adding 6M NH_4OH; boiled on a hot plate for 4 hours to settle down the precipitation and cooled down to room temperature and filtered through the 3.0 μm, 47 mm diameter membrane filter. The precipitate was placed on the planchet, dried under infrared lamp and weighed. Radioactivity was measured in a gross α/β counter of the low background multiple detectors (Berthold LB770).

2.2.3 Standard and Blank Preparation

The calibration standards in coprecipitation were made by adding the standard solution of ^{243}Am dilution to 500 ml of distilled water and adding the standard solution of ^{90}Sr dilution to 2,000 ml of distilled water for gross alpha and beta, respectively, then using the same analytical method described. The blank samples were made using only distilled water and with the same analytical method.

2.3 Study of the Optimum Time, Minimum detectable activity (MDA) and Self Absorption Factor

The sets of samples were made in order to determine the optimum time by being varied from 10-500 minutes, at least twice. The appropriate counting time is the time that so as to obtain a good statistical count by being compared with the standard diviation. The Minimum detectable activity (MDA) were caculated by the equation presented in topic 3 and were compared to each different counting time. The study of self absorption correction factor was already observed by Sarinya *et al.*(2009)[6].

2.4 Study of Mass thickness and Figure of merit (FOM)

The important quantities needed for an estimate of FOM and MDA are the counting efficiency (E) and the background count rate (B). The standard set of this study were prepared as the same method but the carriers were increased proportionally to obtain a reproducible final mass thickness. The range of mass obtained was 20 to 75 mg/cm^2; this range covers all the samples prepared in our laboratory. Four groups of standards of different mass thicknesses were prepared.

3. Calculation

3.1. Efficiency for gross alpha activity was determined by:

$$E = \frac{net\ cpm}{dpm} \quad (1)$$

where E is an Efficiency of detector, net cpm is net count of standard sample and cpm is disintegration rate of standard radionuclide.

3.2. Self-absorption factors can be calculated using the following equation:

$$F_a = \frac{E_x}{E_0} \quad (2)$$

where F_a is the absorption factor, E_x is the mean efficiency of the different points for the curve and E_0 is the mean efficiency of the first point of the curve.

3.3. The trend line of self-absorption factor was quadratic fitting. The function of quadratic fit was determined by:

$$F_a = A \cdot w^2 + B \cdot w + C \quad (3)$$

where A, B and C are the fitting coefficients. The self-absorption curve was determined for each of gross α/β counter of the low background detectors.

3.4. Gross alpha and beta radioactivity can be calculated using the following equation:

$$A = \frac{CPM_s - CPM_b}{60 \cdot E \cdot F_a \cdot V} \quad (4)$$

where F_a is the self-absorption factor, A is the gross radioactivity (Bq/l), CPMs is the averaged count rate of the sample (count per min), CPM_B is the averaged count rate of the blank (count per min), E is the alpha or beta efficiency, V is the volume of the sample aliquot (l) and 60 is the conversion factor from disintegration per minute (dpm) to becquerel (Bq).

3.5. The error activity of measurement was determined by:

$$\sigma = \frac{\sqrt{\frac{cpm_S}{T_S} - \frac{cpm_B}{T_B}}}{60{*}E{*}F_a{*}V} \quad (5)$$

where F_a is the self-absorption factor, δ is error activity of measurement (Bq/l), CPMs is the averaged count rate of the sample (count per min), CPM_B is the averaged count rate of the blank (count per min), E is the alpha or beta efficiency, V is the volume of the sample aliquot (l) and 60 is the conversion factor from disintegration per minute (dpm) to becquerel (Bq).

3.6. The minimum detectable activity was determined by:

$$MDA = \left[\frac{5.29}{60*E*F_a*V}\right]*\sqrt{\frac{cpm_s}{T_s}+\frac{cpm_B}{T_B}} \qquad (6)$$

where *MDA* is the minimum detectable activity in Bq.l–1, T_S is the measuring time of the sample in minutes and T_B is the measuring time of the blank in minutes.

3.7. For low-level activity counting the figure of merit (FOM) is evaluated as follows[7]:

$$FOM = d^2 \frac{E^2 A^2}{4B} \qquad (7)$$

where *d* is the relative uncertainty in the determination of the net count rate produced in the counter by the activity A; B is the background count rate and *E* is the counting efficiency.

The term of figure of merit is used in proportional counting is related with the best compromise between counting efficiency and background. To compare in each set of standard, we assumed that the count of the same activity A, and with the same relative uncertainty d. Thus, FOM can be redefined as[7]:

$$FOM = \frac{E^2}{B} \qquad (8)$$

4. Results and Discussions

4.1 The optimum mass thickness for gross alpha and gross beta counting in sea water sample

The thickness of sample can be effect to efficiency especially in the counting regime of short range radiation such as alpha and beta radiation. As shown in table 1, when the mass of set of standards with same activity was increasing, the efficiency was decreasing. Then it can be assumed that the preparation method of sample that shows low level of mass is an advantage. When compare the coprecipitation technique to standard method (evaporation to dryness) that observed by Suarez-Navarro *et al.* (2001)[4], it can be found that the coprecipitation technique showed signification advantage of final mass of precipitate. The final mass of coprecipitation technique was in the same range and caused no effect with salt. Moreover, the coprecipitation approach also showed several advantages; (i) the use of large volume sample (500 ml for alpha and 2,000 ml for beta sample), (ii) homogeneous of the final precipitate, (iii) appropriate thickness of the final deposit, (iv) sensitive MDA, (v) short measuring time and (vi) high reproducibility of the final weight of the residue[3, 4, 6]. Moreover, the content of initial volume of sample can be control.

Table 1 The mass thickness of each set of standard preparing by coprecipitation technique

Set of sample									
Detecter	CPM_B	CPM_S				Efficiency			
		1x	2x	3x	4x	1x	2x	3x	4x
1	0.0900	2.1600	1.3000	1.0500	0.6700	0.1568	0.0917	0.0727	0.0439
2	0.0700	2.0600	1.1700	0.8400	0.5200	0.1508	0.0833	0.0583	0.0341
3	0.0500	1.7200	1.1200	0.7200	0.5900	0.1265	0.0811	0.0508	0.0409
4	0.0900	2.1300	0.9400	0.8500	0.4600	0.1545	0.0644	0.0576	0.0280
5	0.0500	1.9100	1.0100	0.7900	0.5900	0.1409	0.0727	0.0561	0.0409
6	0.1200	2.2700	1.1500	1.0200	0.6900	0.1629	0.0780	0.0682	0.0432
7	0.0800	2.2500	1.1400	0.9300	0.7300	0.1644	0.0803	0.0644	0.0492
8	0.0800	2.1700	1.2300	0.9000	0.7400	0.1583	0.0871	0.0621	0.0500
9	0.2200	1.9800	0.9100	0.6900	0.7400	0.1333	0.0523	0.0356	0.0394
10	0.1300	2.1000	1.1400	0.9100	0.7400	0.1492	0.0765	0.0591	0.0462

Note: x = double time of carrier

4.2 The self absorption factor for gross alpha and gross beta counting in sea water sample[3, 6]

The standards for self absorption correction factor were prepared by adding the solution of ^{243}Am diluted to 500 ml of distilled water and varying the amount of the carriers used in the process to increase the weight of the final precipitate. The amounts of carriers used in the first standard were the same as those used in the radiochemical method. The carriers were increased proportionally to obtain a reproducible final weight. The range of weight obtained was 20 to 75 mg; this range covered all the samples prepared in our laboratory. Four groups of standards of different weights were used in this study. Figure 9 shows the self-absorption correction factor (Fa) as a function of the precipitate weight for gross α/β counter (Berthold LB770). No criteria were found in the literature about the fitting function. In this case of study the trend line of self-absorption factor was quadratic fitting. The functions of quadratic fit were determined by equation 3 and calculated to the result that showed in equation 9.

The self-absorption factor was calculated for different weights of the final precipitate using the equation of each detector. The average equation can be used:

$$F_a = 0.0003w^2 - 0.0414w + 1.692 \qquad (9)^{3,6}$$

where F_a is the self-absorption factor and w is the weight of the final precipitate mass in milligrams.

The standards for self absorption correction factor were prepared by adding the solution of ^{90}Sr diluted to 2000 ml of distilled water and varying the amount of the carriers used in the process to increase the weight of the final precipitate. The amounts of carriers used in the first standard were the same as those used in the radiochemical method. The carriers were increased proportionally to obtain a reproducible final weight. The range of weight obtained was 0.5 to 50 mg; this range covers all the samples prepared in our laboratory. Four groups of standards of different weights were used in the study. The results showed that the self absorption factor cause no effect with gross beta counting[3, 6]

4.2. The mass thickness and figure of merit (FOM) of detection regime

The term figure of merit is related with the best compromise between counting efficiency and background in low level activity. As mentioned before, the important factor needed for an estimate of FOM and MDA are the counting efficiency (E) and the background count rate (B)[7]. When considered with efficiency, it can be assumed that the mass thickness was involved. In figure 1, the FOM decreased when mass thickness increased. It was shown that the compromise between counting efficiency and background in low level counting were decreased following by the increasing of mass thickness. Then it can be assumed that the mass and thick of sample was the important parameter for gross counting in short range radiation. Even though this problem can be solved by self absorption correction factor but it cannot estimate the great result if the groups of final precipitation are significant difference.

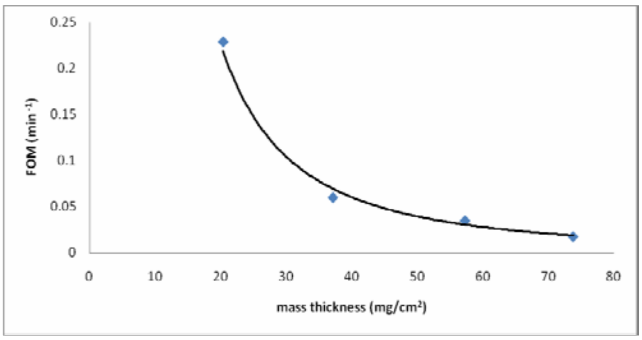

Figure 1 The figure of merit and mass thickness of sample

4.3. The optimum time and Minimum detectable activity (MDA) for gross alpha and gross beta counting in sea water sample

The group of sea water samples was collected from Chon Buri province, the Upper Gulf of Thailand. The samples were prepared following to its method and counted, varying the counting time from 10 minutes to 500 minutes. The result shows that the standard deviation was decreasing as counting time increasing and the optimum counting statistic errors were shown from 100 minute for gross alpha (Figure 2) and 200 minute for gross beta (Figure 3). At that counting time, the results of measurement also showed the suitable of minimum detectable activity (MDA). Then the selected counting time for gross alpha and gross beta in sea water samples in this experiment had been used at 100 minute and 200 minute, respectively.[3, 6]

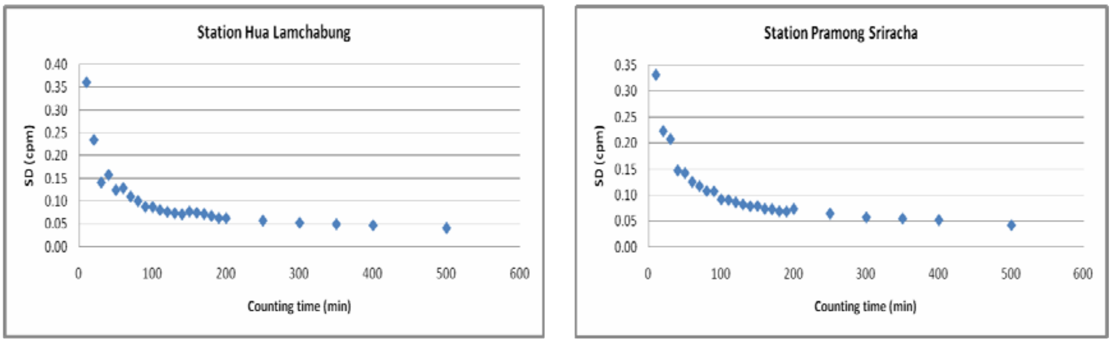

Figure 2 Range of standard deviation of gross alpha counting in sea water sample, the measuring times varied from 10 to 500 minute

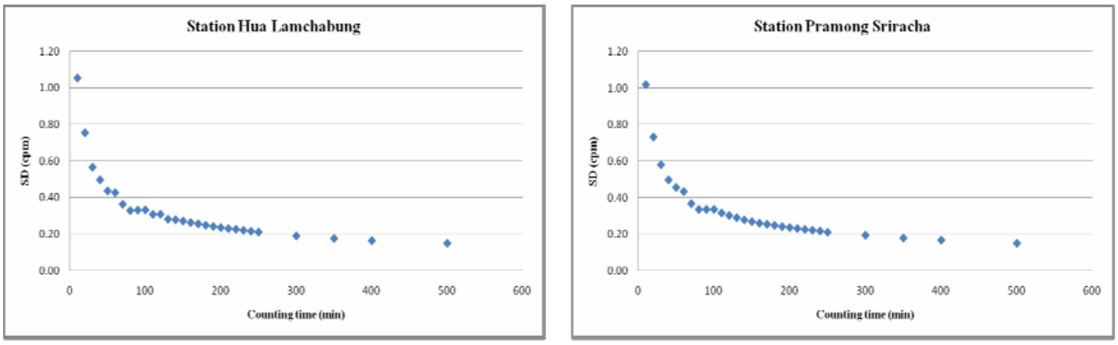

Figure 3 Range of standard deviation of gross beta counting in sea water sample, the measuring times varied from 10 to 500 minute

5. Summary

A statistical study of the detection process demonstrates that the free parameter is essential to compute the counting efficiency. The optimizations of each basic parameter of measurement such as optimum time, mass and thickness can be estimated to the great result of measurement system. It has been shown that with the method described it is possible to assess how close to the optimal sampling and counting regime for a specific radionuclide is an existing procedure in environmental monitoring of fresh and seawater. Also, when developing a monitoring program, on the basis of the method presented, it is possible to plan the sampling, preparation method and counting regimes and to assess the minimum detectable activities that can be attained.

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7. References

1. Environmental Protection Agency (EPA). 2002. Guidance on Choosing a Sampling Design for environmental Data Collection. Office of environmental information, Washington.
2. International Atomic Energy Agency (IAEA). 1997. Strategies and Methodologies for Applied Marine Radioactivity Studies. Training Course Series No. 7.
3. Sarinya Wongsanit, Pannee Pakong, Yutthana Tomnoi and Suchin Udomsomporn 2010. Determination of Gross Alpha and Beta Activities in Seawater from the Upper Gulf of Thailand Using Coprecipitation Technique. 2010. *In* the third Asian and Oceanic Congress on Radiation Protection (AOCRP-3).

4. J.A. Suarez-Navarro, LI. Pujol and M.A. de Pablo. 2001. Rapid determination of gross alpha-activity in sea water by coprecipitation. Journal of Radioanalytical and Nuclear Chemistry. 253: 47-52.
5. N. Damla, U. Cevik, G. Karahan and A.İ. Kobya. 2006. Gross α and β activities in tap waters in Eastern black Sea region of Turkey. Journal of Chemosphere. 62: 957-960.
6. Sarinya Wongsanit, Pannee Pakong, Yutthana Tomnoi and Suchin Udomsomporn. 2009. The Optimum Conditions for the Determination of Gross-Alpha Radioactivity in Seawater by Coprecipitation Technique. 2009. *In* ^{7}th Eco-Energy and Materials Science and Engineering Symposium, Chiang Mai, Thailand 17-20 November 2009. Tokyo, Japan 24-28 May 2010.
7. F. Vaca, G. Manjion, S. Cuiellar, M. Garcıa-Leon. 2001. Factor of merit and minimum detectable activity for ^{90}Sr determinations by gas-flow proportional counting or Cherenkov counting. Applied Radiation and Isotopes 55: 849–851.