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A STUDY ON THE SIMULTANEOUS MEASUREMENTS OF BETA EMITTING ISOTOPES

Goung Jin Lee, Seoung Pyung Kim
Chosun University, College of Engineering, Department of Nuclear Engineering
P.O. 501-759, #375, Seosuk-Dong, Gwangju, Republic of Korea

Abstract - Beta radiation is measured for an environmental monitoring purpose or for an internal radiation exposure monitoring of nuclear power plant's worker. In korea, strontium 89 and strontium 90 is measured for an environmental monitoring purpose. Also tritium and carbon 14 contained in urine is measured for an internal radiation exposure monitoring of nuclear power plant's worker. Because above isotopes emits low energy beta radiations having a wide range of energy, very complicated isotope separation preprocess is needed. In this study, two mixed beta emitting isotopes are measured simultaneously using a liquid scintillation counter(LSC) and analyzed by using a developed statistical method. Banded least square method is used to analyze the mixed spectrum, and the goodness-of-fitness test is proposed. Test results show that the developed procedure can be very useful for analyzing a mixed beta emitting isotopes.

INTRODUCTION

1. Research Background

Because radioactive beta decay isotopes emits low energy beta radiations having a wide range of energy, very complicated isotope separation preprocess is needed before measuring it. Up to now^{89}, Sr and ^{90}Sr is separated using a series of complicated chemical processes[1~3]. In case of ^{3}H and ^{14}C in urine, discrimination window is used to determine the existence of C^{14} in urine. If there exists meaningful amount of C^{14} in urine, a complicated chemical separation process is used to separate ^{3}H and ^{14}C[4]. So the overall pre-treatment procedure to measure ^{89}Sr/^{90}Sr or ^{3}H/^{14}C is too complicated. In this study, an analytical procedure to analyze multiple beta emitting isotopes simultaneously is developed to simplify the chemical separation procedure.

2. State of Art ^{89}Sr/^{90}Sr Measurement Method

In korean nuclear power plants, ^{89}Sr/^{90}Sr is measured for an environmental monitoring purpose. Both of the ^{89}Sr and ^{90}Sr is produced either from nuclear bomb test or from nuclear power generation. But because the nuclear bomb tests were ended more than 20 years ago, due to the short decay half life of ^{89}Sr, remaining ^{89}Sr produced from the nuclear bomb test is below the MDA level.

If a measured ratio of environmental ^{89}Sr to ^{90}Sr is very low, it can be concluded that the measured radioactive strontium is from the former nuclear bomb tests. On the other hand, if the measured ratio of ^{89}Sr to ^{90}Sr is high, the measured radioactive strontium is either from a nuclear power plant or from a recent nuclear accident.

Because ^{89}Sr and ^{90}Sr gives wide range of weak energy beta spectrum, state of art ^{89}Sr/^{90}Sr measurement process is very complicated, as follows[1~3].

- Process 1 : Separation of strontium from the other intervening isotopes
- Process 2 : Wait until ^{90}Y growth
- Process 3 : Measure total activity of ^{89}Sr + ^{90}Sr + ^{90}Y
- Process 4 : Separation of ^{90}Y from strontium
- Process 5 : Measure activity of ^{90}Y
- Process 6 : Calculation of activity of ^{90}Sr and ^{89}Sr

The state of art $^{89}Sr/^{90}Sr$ measurement procedure is too complicated, expensive, time consuming, and have high possibility of much error.

3. State of Art $^3H/^{14}C$ Measurement Method

^3H and/or ^{14}C is a potential sources of internal exposure of nuclear power plant workers. So, ^3H and/or ^{14}C in Urine is measured for an internal exposure estimation of nuclear power plant workers. Because a simultaneous measurement procedure of $^3H/^{14}C$ was not developed, the state of art $^3H/^{14}C$ measurement process is very complicated, as follows[4].

- Set discrimination window of ^{14}C
- Process 1 : Measure total activity of ^3H and ^{14}C
- Process 2 : If there exists meaningful amount of ^{14}C, chemical separation of $^3H/^{14}C$ is performed
- Process 3 : Measure ^3H and ^{14}C separately

ANALYTIC SPECTRUM SEPARATION METHOD

1. Least Square Method

To fit a measured data to a reference data, usually least square method is used. In case of an analytic separation of mixed spectrum made of two components, the sum of square errors (SSE) of counts between the measured spectrum and the modeled spectrum is given as follows[5],

$$SSE = \sum_{i=1}^{channel} [F_{measured,i} - (\alpha_A \cdot F_{A,i} + \alpha_B \cdot F_{B,i})]^2 \quad (1)$$

where

$F_{measured,i}$: measured counts in channel i of a sample,

$F_{A,i}$: reference counts in channel i of A-isotope of unit activity,

$F_{B,i}$: reference counts in channel i of B-isotope of unit activity,

α_A : activity of A-isotope (to be Calculated),

α_B : activity of B-isotope (to be Calculated),

i : channel index ($i = 1, 2, \cdot \cdot, Nband$).

To minimize SSE, equation(1) is partially differentiated with respect to α_A and α_B.

$$\frac{\partial SSE}{\partial \alpha_A} = 0 \text{ and } \frac{\partial SSE}{\partial \alpha_B} = 0 \quad (2)$$

This gives,

$$\alpha_A \sum_{i=1}^{Channel} F_{A,i}^2 + \alpha_B \sum_{i=1}^{Channel} F_{A,i} \cdot F_{B,i} = \sum_{i=1}^{Channel} F_{A,i} \cdot F_{measured,i} \quad (3)$$

and

$$\alpha_A \sum_{i=1}^{Channel} F_{A,i} \cdot F_{B,i} + \alpha_B \sum_{i=1}^{Channel} F_{A,i}^2 = \sum_{i=1}^{Channel} F_{B,i} \cdot F_{measured,i} \quad (4)$$

Activity of A isotope(α_A) and B isotope(α_B) can be calculated algebraically by combining equation(3) and (4).

In case of radiation energy spectrums, there are random fluctuations among channels due to the randomness of radioactive decay. So the usual least square method doesn't work.

2. Banded Least Square Method

To handle the random fluctuations of spectrums, the spectrums are smoothened by grouping the energy channels into 4 ~ 10 bands.

When using liquid scintillation counter(LSC) having 1024 energy channels, in case of $^3H/^{14}C$ analysis the most of counts are from 1~450 channel. In stead of using the originally measured spectrum, we transformed it into a banded spectrum(fig 1) using a 7 band given in table 1.

Table 1. BAND for ^3H, ^{14}C Analysis

BAND 1	1~50 Channel
BAND 2	51~100 Channel
BAND 3	101~150 Channel
BAND 4	151~200 Channel
BAND 5	201~300 Channel
BAND 6	301~400 Channel
BAND 7	401~500 Channel

Using the transformed banded spectrums, banded least square procedure is used to give following results,

$$\alpha_A \sum_{i=1}^{Nband} F_{A,i}^2 + \alpha_B \sum_{i=1}^{Nband} F_{A,i} \cdot F_{B,i} = \sum_{i=1}^{Nband} F_{A,i} \cdot F_{measured,i} \quad (5)$$

and

$$\alpha_A \sum_{i=1}^{Nband} F_{A,i} \cdot F_{B,i} + \alpha_B \sum_{i=1}^{Nband} F_{A,i}^2 = \sum_{i=1}^{Nband} F_{B,i} \cdot F_{measured,i} \quad (6)$$

3. A Goodness-of-Fit Test

Even though there are several types of goodness-of-fit tests[5], in this paper root mean square errors (RMSE) between the measured banded spectrum and the model spectrum is used as follows,

Root Mean Square Errors =

$$\sqrt{\sum_{i=1}^{Nband} \frac{(f_{i,ref} - f_{i,measurement})^2}{Nband}} \quad (7)$$

where

$f_{i,measurement}$: normalized measured banded spectrum of a sample,

$f_{i,ref}$: normalized spectrum obtained from reference banded spectrums,

i : band index (i = 1, 2, · · , Nband).

Very small value of calculated root mean square errors(RMSE << 1) means that there are no intervening isotopes exists and spectrum separation works.

EXPERIMENTS

1. $^{89}Sr/^{90}Sr$ Experiments

Using the developed banded least square method, following simultaneous $^{89}Sr/^{90}Sr$ measurement procedure was proposed.

- Process 1 : Separation of strontium from the other intervening isotopes
- Process 2 : Wait until ^{90}Y growth
- Process 3 : Measure total spectrum of $^{89}Sr + ^{90}Sr + ^{90}Y$
- Process 4 : Separation of mixed spectrum using the developed analytic method

To validate and verify the developed mathematical spectrum separation procedure, various ratio of mixed $^{89}Sr/^{90}Sr$ sample having low level activity was analyzed as shown in Fig. 2 and table 2.

2. $^{3}H/^{14}C$ Experiments

For the simultaneous measurement $^{3}H/^{14}C$, following procedure was proposed.

- Process 1 : Measure total spectrum of ^{3}H and ^{14}C
- Process 2 : Separation of mixed spectrum using the developed analytic method

Various test results are shown in Fig. 3 and table 3.

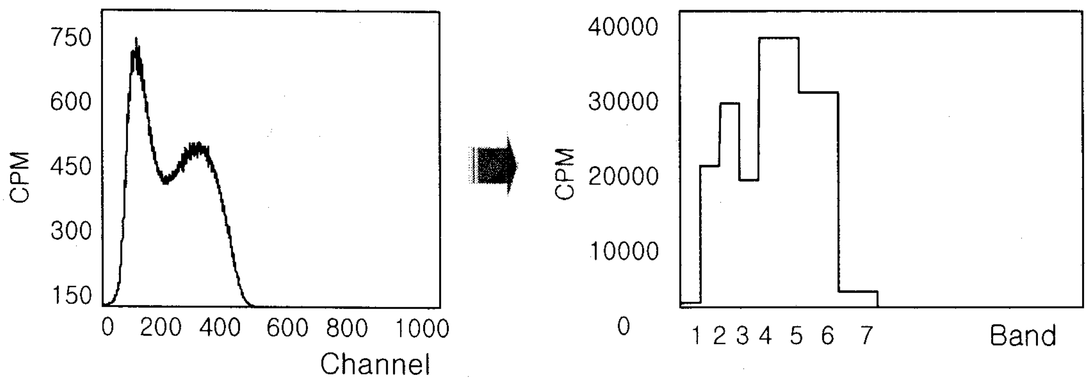

Fig. 1. Transformation of Measured Spectrum into Banded Spectrum

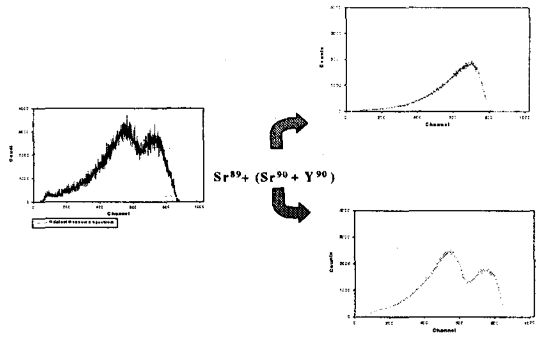

Fig. 2. Example of $^{89}Sr/^{90}Sr$ Separation

Table 2. Experimental Results of Simultaneous Analysis of $^{89}Sr/^{90}Sr$

	Mixing Ratio	Labeled Activity(Bq)		Analyzed Activity(Bq)		Ralative error(%)		RMSE
	^{90}Sr : ^{89}Sr	^{90}Sr	^{89}Sr	^{90}Sr	^{89}Sr	^{90}Sr	^{89}Sr	
1	1 : 1.6	4.70	7.62	4.54	7.71	-3.48	1.21	0.374E-02
2	1 : 3.2	4.76	15.40	4.74	15.50	-0.32	0.26	0.382E-02
3	1 : 7.4	4.55	33.60	4.46	33.20	-1.98	-1.02	0.774E-02
4	1 : 15.0	4.67	69.50	5.17	68.70	10.70	-1.27	0.143E-01

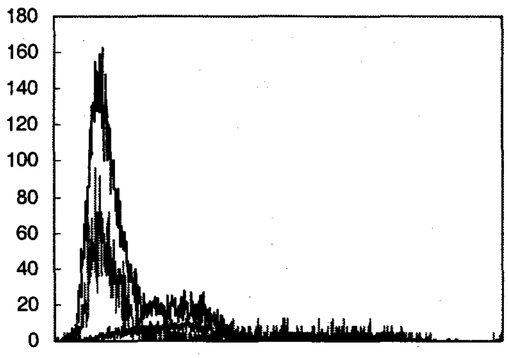

Fig. 3. Example of $^{3}H/^{14}C$ Separation

Table 3. Experimental Results of Simultaneous Analysis of $^{3}H/^{14}C$

	Mixing Ratio	Labeled Activity(Bq)		Analyzed Activity(Bq)		Relative error(%)		RMSE
	^{3}H : ^{14}C	^{3}H	^{14}C	^{3}H	^{14}C	^{3}H	^{14}C	
1	13 : 1	30.2	2.8	28.6	2.6	-5.6	-7.7	0.470E-02
2	3 : 1	6.1	2.4	6.7	2.3	+8.9	-4.3	0.512E-02
3	1 : 1.5	3.1	2.4	3.5	2.2	+12.9	-9.0	0.767E-02
4	1 : 4	6.3	25.3	6.5	23.8	+3.1	-6.0	0.646E-02
5	1 : 10	6.3	83.5	8.6	85.9	-30.0	-2.8	0.121E-01

RESULTS AND CONCLUSIONS

1. Results

Very low activity level of $^{3}H/^{14}C$ and $^{89}Sr/^{90}Sr$ mixture sample was measured and analyzed. Sample was measured for 30 minutes using Wallac Guardian 1414. In case of $^{3}H/^{14}C$ analysis, the relative error for ^{3}H and ^{14}C is kept lower than 15% and 10%, respectively. In case of $^{89}Sr/^{90}Sr$ analysis, the relative error for ^{89}Sr and ^{90}Sr is kept lower than 10%.

Goodness-of-fit test shows that any of two mixed spectrum can be separated very well.

2. Conclusions

A mathematical spectrum separation procedure was developed, and computer code package to analyze mixed spectrum of LSC was developed. Various test results shows any of two mixed β spectrums can be separated and analyzed using the developed banded least square method.

Developed method can be used for β ray monitoring applications.

Advantages of the developed mathematical spectrum separation process can be summarized as,

- Very accurate
- Easy
- Economical
- Time Saving

due to simplified chemical pre-treatment process.

Another advantage is existence of unwanted intervening isotopes in a sample can be easily recognized using goodness-of-fit test.

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