

# Application Note

Alpha/Beta

ABA-007

## Determination of Alpha Radioactivity in Vegetable Ashes With Liquid Scintillation Analysis

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### Abstract

A rapid method for the determination of alpha activity in vegetable ashes using alpha/beta liquid scintillation analysis (LSA) has been studied. After ashing vegetable samples in a muffle furnace, 50-200 milligrams of ash was dissolved in a 20 mL glass counting vial with  $\text{HNO}_3$  and  $\text{H}_2\text{O}_2$ . This solution was then mixed with diisopropylnaphthalene (DIN)-based cocktail, Ultima Gold® AB and F. With an optimum pulse decay discriminator (PDD) setting, the counting efficiency of alpha emitters is nearly 100% and the minimum detectable concentration is as low as  $4.5 \times 10^{-2}$  Bq/kg (fresh).

### Introduction

Assessment of any release of radioactivity to the environment is important for the protection of public health, especially if the released activity can enter the food chain. Such assessment demands a rapid, reliable and practical measurement technique.<sup>1</sup> It is known that low level gamma spectrometry is a suitable method for the assay of gamma emitting nuclides in environmental samples if a sufficient quantity of sample is collected. Both qualitative and quantitative information about gamma emitting nuclides can be obtained. However, for the assay of alpha emitting nuclides in the environment, the task is much more complicated than that for gamma emitting nuclides. Because of the possible absorption of alpha particles by the physical matrix, long chemical separations followed by electrodeposition are usually needed. In the event of accidental releases of radioactivity, it is essential for the radiation protection authorities to assess possible contamination. The methods mentioned above are obviously not satisfactory for the rapid screening of alpha emitting nuclides.

Our recent work with LSA showed that our methodology seems to be a possible alternative for the environmental monitoring of alpha activity. Since the samples are homogeneously dispersed in the scintillator medium, the problems of sample self-absorption are avoided and a high detection efficiency can be obtained. Another important advantage of LSA is its rapid and convenient sample preparation. The major problem with a traditional LSA was the high background produced by beta activity, which is usually much higher than alpha activity. With the development of pulse discrimination electronics, such as pulse decay analysis (PDA), as well as cocktails designed for optimum alpha/beta separation, the interference between alpha and beta events can be greatly reduced and the alpha background decreased by a factor of ten. All of these recent developments provide the capability to determine alpha emitting nuclides with LSA.

The purpose of this work was to test and evaluate the PDD function and new cocktails for the determination of alpha activity in vegetable samples with LSA.

### Experimental

Instrumentation: Tri-Carb® 2550TR/AB liquid scintillation analyzer, Packard Instrument Company

Scintillation cocktails: Ultima Gold AB and Ultima Gold F, Packard Instrument Company

### Procedure

Chinese cabbage, carrot, and potato were collected as the vegetable samples. The sample preparation procedures follow:



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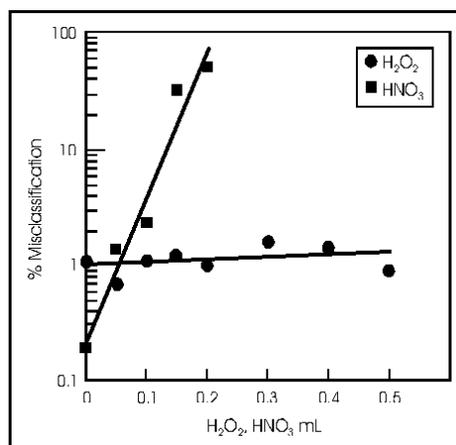
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1. Wash the samples with water.
2. Dry the material at room temperature for 24 hours and then in a hot blast stove at a temperature of 100-105 °C for 24 hours.
3. Put the dried samples into a muffle furnace at a temperature of 600 °C for three to four hours, until no carbon remains.
4. Weigh 50-200 milligram of ash (about 10-50 gram fresh vegetable) in a 20 mL glass counting vial.
5. Add 0.75 mL of 30% H<sub>2</sub>O<sub>2</sub> and 0.15 mL of concentrated HNO<sub>3</sub>.
6. Heat the vial gently on a hot plate.
7. After all of the ash is dissolved, continually heat to dryness.
8. Dissolve the residue with 0.5 mL of H<sub>2</sub>O<sub>2</sub> and 0.05 mL of concentrated HNO<sub>3</sub> by gentle heating.
9. Mix the solution with 5 mL of Ultima Gold AB and then add 10 mL Ultima Gold F.
10. Measure the sample in a liquid scintillation analyzer with a PDD setting of 95-110 which is optimal for this particular instrument and cocktail matrix.



**Figure 1.**  
Percent misclassification of plutonium vs. the amount of H<sub>2</sub>O<sub>2</sub> and HNO<sub>3</sub> at a constant PDD.

## Results and Discussion

### Pretreatment of the Ash

The ash has to be pretreated with H<sub>2</sub>O<sub>2</sub> and concentrated HNO<sub>3</sub>. The amount and ratio of H<sub>2</sub>O<sub>2</sub> and HNO<sub>3</sub> should be properly chosen to dissolve all of the ash with a minimum amount of concentrated HNO<sub>3</sub>. Figure 1 shows the influence of the amount of H<sub>2</sub>O<sub>2</sub> and HNO<sub>3</sub> in the cocktail on the misclassification of <sup>239</sup>Pu and <sup>240</sup>Pu, which were used as the alpha radionuclides in this procedure. Excess HNO<sub>3</sub> in the cocktail results in an increase in misclassification. From our experiments, 0.5 mL of 30% H<sub>2</sub>O<sub>2</sub> and 0.05-0.1 mL of concentrated HNO<sub>3</sub> are the optimum amounts for the dissolution of the ash residue.

### Amount of Ash

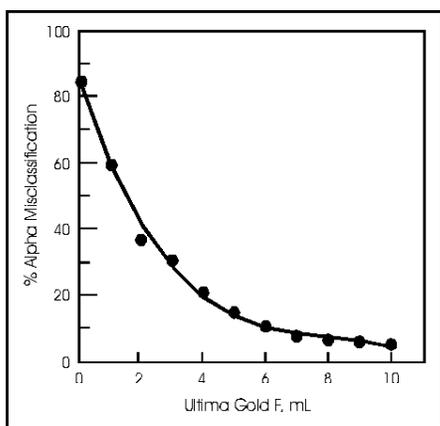
Table 1 shows the influence of the amount of ash on the recovery of plutonium. Up to 200 mg ash can be mixed with the cocktail mixture of Ultima Gold AB (UGAB) and F (UGF) in a 20 mL glass vial for direct counting. This amount of ash corresponds to about 40-50 g of fresh vegetables. Our experiments showed that adding more than 200 mg of ash into a vial could produce a non-homogeneous solution of ash and cocktail.

### Cocktail Comparison

Different cocktail formulations have been tested and compared. Traditional formulations for beta counting, dioxane or xylene-triton-based cocktails, do not perform as well as di-isopropylnaphthalene (DIN)-based cocktails, such as Ultima Gold AB and Ultima Gold F. The main difference between Ultima Gold AB and Ultima Gold F is that the former contains emulsifiers and the later does not. Figure 2 shows that better alpha/beta separation can be achieved as the ratio of Ultima Gold F increases. A smaller amount of emulsifier in the cocktail produces better alpha/beta separation and lower alpha background.

Ash (mg)		Pu-239 & 240		
		Added (DPM)	Measured (DPM)	Recovery %
53	cabbage	238	236	99
106	cabbage	238	235	99
159	cabbage	238	234	98
265	cabbage	238	194	82
220	carrott	238	236	99
264	carrott	238	238	100
170	potato	238	223	95
255	potato	238	178	76

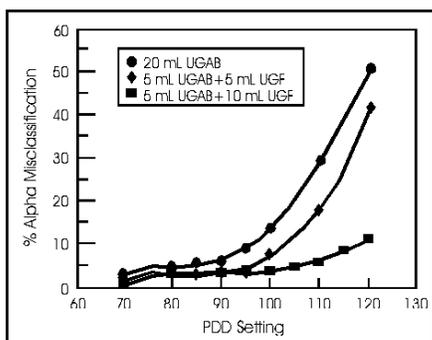
**Table 1.**  
Influence of amount of ash on recovery of plutonium.



**Figure 2.**

Percent misclassification vs. the amount of Ultima Gold F (Pu-ash, 5 mL Ultima Gold AB and PDD setting 90).

Figure 3 shows the influence of different ratios of Ultima Gold AB and F on the misclassification at various PDD settings. For optimum results use 5 mL Ultima Gold AB and 10 mL Ultima Gold F. Figure 4 shows the influence of cocktail composition on alpha background at various PDD settings.



**Figure 3.**

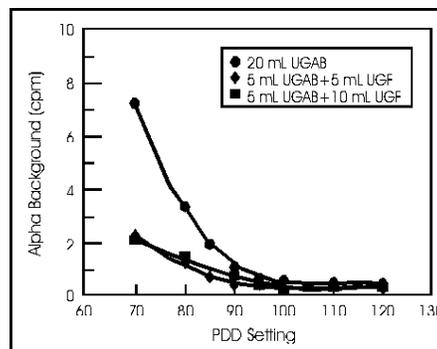
Alpha misclassification vs. PDD setting with different cocktail composition.

### PDD Setting

In order to acquire good alpha/beta separation, a proper PDD setting is necessary. With earlier model Tri-Carb LSAs, specially modified alpha/beta systems, the operator had to adjust the PDD setting manually with two reference samples, one pure beta emitting sample and one pure alpha emitting sample. With the newer model Tri-Carb 2550TR/AB, the instrument can determine the optimum PDD setting automatically. Figure 5 shows the PDD setting plot acquired by the instrument, in which both the misclassification of alpha and beta events at the different PDD settings and the optimum (intersection of two curves) are given. Table 2 compares the percent alpha misclassification of the manual method (early model alpha/beta LSA) to the automatic method at individual PDD settings. They are quite similar.

The PDD setting can be determined automatically by the Tri-Carb 2550TR/AB as follows:

1. Prepare two ash samples as described in the Analytical Procedure section, steps 1-4.
2. Add 5-10  $\mu\text{L}$   $^{90}\text{Sr}/^{90}\text{Y}$  into the first vial and 5-10  $\mu\text{L}$   $^{241}\text{Am}$  into the second one. (>50,000 DPM each)
3. Add cocktail as described in steps 5-6 (Analytical Procedure section).
4. Measure the samples in a Tri-Carb 2550TR/AB LSA as alpha/beta standards. The instrument will compute the optimum PDD setting.

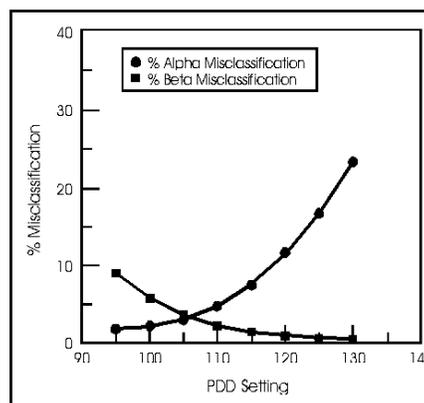


**Figure 4.**

Alpha background vs. PDD setting with different cocktail composition.

### Analytical Procedure

1. Weigh 50-200 mg of vegetable ash in a 20 mL glass counting vial.
2. Add 0.75 mL of 30%  $\text{H}_2\text{O}_2$  and 0.15 mL of concentrated  $\text{HNO}_3$ .
3. Heat the vial gently on a hot plate until all the ash is dissolved. Continue to heat to dryness.
4. Dissolve the residue with 0.5 mL of 30%  $\text{H}_2\text{O}_2$  and 0.05 mL of concentrated  $\text{HNO}_3$ .
5. Add 5 mL of Ultima Gold AB and mix the cocktail with the ash solution completely.
6. Add 10 mL of Ultima Gold F.
7. Measure the sample in a liquid scintillation analyzer with an optimum PDD setting.



**Figure 5.**

Misclassification vs. PDD setting.

PDD	% Alpha Misclassification	
	Manual Method	Automatic Method
90	3.6	--
95	4.3	1.7
95	4.1	2.2
105	5	3
110	6.2	4.6
115	8.8	7.2
120	11.9	11.1

**Table 2.**

Comparison of the results with manual and automatic method to determine the optimum PDD setting.

No.	Pu-239 & 240		
	Added (DPM)	Measured (DPM)	% Recovery
1	74.2	73	98.4
2	74.2	74.2	100
3	74.2	72.3	97.4
4	74.2	77	103.8
5	74.2	74.4	100.3
6	74.2	73.2	98.6
Average	74.2	74.0±1.7	99.7±2.3%

**Table 3.**

Recovery of plutonium with LSA.

### Recovery and minimum detectable activity

Table 3 shows the recovery of the method. By adding 1.23 Bq <sup>239+240</sup>Pu into the 106 mg vegetable sample, the recovery was found to be 99.7±2.3% (n=6).

The minimum detectable activity (MDA) can be expressed by<sup>6</sup>:

$$\text{MDA (unit)} = \frac{4.65Sb+2.71}{K}$$

Where 4.65 is derived for a 5% probability of a type I and type II errors for a paired blank and sample measurement. Counts for the zero blank case are 2.71. K represents a series of factors.

$$K = \frac{(\text{activity units}) (\text{efficiency}) (\text{aliquot})}{(\text{abundance}) (\text{attenuation}) (\text{counting time}) (\text{chemical yield}) (\text{decay correction})}$$

Sb is defined as the standard deviation of the total counts of an appropriate blank, which is a sample identical in physicochemically and radiologically significant ways to the sample to be analyzed, except that it contains no quantity of the analyte to be measured, according to ANSI N13.30.<sup>7</sup> The factors which tend to influence minimal detectable activity limits are the counting efficiency, the quantity of the sample, the counting time and the background. To obtain low MDAs, the counting efficiency should be high and the background should be as low as possible. Doubling the counting efficiency improves the detection limits by a factor of two, but reducing the background by half can only improve the detection limits by a factor of the square root of two (2<sup>1/2</sup>). The counting efficiency for alpha emitters with LSA is nearly 100% and the observed alpha background in this experiment is 0.0033 CPS (0.2 CPM) with a PDD setting of 105 for an optimum alpha region of interest. Therefore, the MDA of the method can be estimated to be 4.5 x 10<sup>-2</sup> Bq/kg (fresh) or 8.4 x 10<sup>-6</sup> Bq/mg (ash) with 160 mg vegetable ash (about 30 g fresh vegetable) and 900 minutes counting time.

### Conclusion

By using a DIN-based cocktail, such as Ultima Gold AB and F, and the PDA feature, up to 200 mg vegetable ash can be determined directly with liquid scintillation analysis. The Minimum Detectable Activity (MDA) is 4.5 x 10<sup>-2</sup> Bq/kg (fresh) with 160 mg vegetable ash, 900 minutes counting time at a PDD setting of 105. This method is simple, rapid and sensitive for the monitoring of alpha activity in environmental vegetation.

### References

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