Application Note

Alpha/Beta ABA-001

Gross Alpha Counting Of Air Filters Using A Pulse-Shape Discriminating Alpha Liquid Scintillation Counter

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The Health Physics Department at the Oak Ridge Y-12 Plant collects ~120,000 air samples from 1,016 locations in the Oak Ridge Y-12 Plant each year for gross alpha analysis. The sampling locations contain a computer card, which is prepunched with the location-specific data, and an integral filter window through which several cubic meters of air are pulled. The cards are collected either daily or weekly, and the Plant Laboratory receives ~500 samples per day for analysis.

The samples have been routinely analyzed on two in-house fabricated, automated alpha counters, which used automated card readers to decode the punched information and silicon surface-barrier detectors for one minute counts. The low-counting efficiency (~30%), a large sample absorption factor (~30%), and high maintenance requirements on the card readers have been major problems.

DOE Order 5480-11 lowered respirator limits from 220 DPM/m^3 to 44 DPM/m^3. Because the Y-12 Plant Action Value is set at 0.1 of the requirements, the limit was thereby lowered to 4.4 DPM/m^3 within the plant. The low counting efficiency and the uncertainty associated with the high absorption factor made the existing counting systems unsuitable for counting samples under the new limit. An interim procedure that uses windowless gas flow proportional counters was adopted to comply with the new DOE order (the procedure is shown in Figure 1). Five percent of the samples are internal controls. The controls are plotted on control charts, and a recovery factor (90%) is calculated from the average recovery of the controls and then applied to the final answers. The data are electronically transferred to the customer daily.

The procedure has the following advantages: (1) maintaining the required sample throughput of 500 samples plus 25 controls; (2) higher counting efficiency (50%); and (3) lower detection limits; however, the procedure is labor intensive (four people are required). To reduce the labor requirements, another technique (pulse decay discriminating alpha liquid scintillation counting) was identified as a possible automated replacement for the existing method. Within the last year, the Packard Instrument Company has built alpha/beta LSC instruments for use in Europe. The Y-12 Plant purchased the first three instruments sold in the U.S. The instruments combine the advantages of high counting efficiency (~100% for alpha) and high throughput of samples. This paper will describe the evaluation of the instrument for gross alpha analysis of air filters.

A study was performed to determine the alpha efficiency of the scintillation counter. Twenty-five microliters each of several enriched uranium standards were added directly into 20 mL of scintillation cocktail (InstaGel™ XF/20% naphthalene; Packard Instrument Company), and a 15 minute count was obtained for each vial. The results are shown in Table 1.

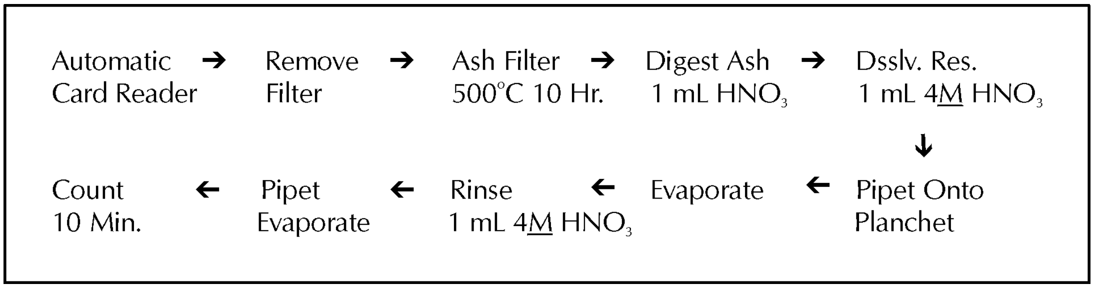

Figure 1.
Interim Procedure Using Windowless Gas Flow Proportional Counters.

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Standard DPM	CPM Scintillation	Percent Efficiency
0	3	---
32	36	104
63	70	105
126	130	100
319	316	98
623	690	110
967	991	102
1297	1326	102
1641	1725	105
		Average 103+/- 3

Table 1.
Alpha Efficiency of Scintillation Counter.

A study was performed to determine the alpha efficiency of the scintillation counter. Twenty-five microliters each of several enriched uranium standards were added directly into 20 mL of scintillation cocktail (InstaGel™ XF/20% naphthalene; Packard Instrument Company), and a 15 minute count was obtained for each vial. The results are shown in Table 1.

The original intent of using the instrument was to gross alpha count air filters directly in the scintillation cocktail, with no preparation. A second test was performed to check the validity of this idea. Fifteen "real" samples were alpha counted by using the existing automated air counters. The samples were then spiked with 967 DPM of enriched uranium and counted for 15 minutes in the scintillation counter. The results in Table 2 indicate an approximate loss in efficiency of >50%. Obviously, counting the filters directly is not a very efficient technique. A test was performed in an attempt to determine what effect the position of the filter in the vial had on the count rate. A vial was selected that contained a dirty filter spiked with 967 DPM alpha. The filter was oriented such that the concave portion of the filter faced the back of the instrument (0 position) and then counted. The vial was rotated 90, 180, and 270 degrees and counted at each position. The data indicate that the filter position has little effect on the count (as shown in Table 3).

Another experiment was performed with clean and dirty filters to determine the effect of the filter paper on the count rate. A total of 1641 DPM alpha was spiked directly into the cocktail contained in vials with clean and dirty filters. As Table 4 indicates, there was a slight drop in efficiency due to the black spot on the filter paper, although the filter paper itself had no effect. Therefore, it was concluded that the major problem was the alphas embedded in the filter paper or in the dirt.

Original DPM	Spike DPM	Percent Efficiency	Original DPM	Spike DPM	Percent Efficiency
461	967	48	170	967	18
518	967	54	192	967	20
426	967	44	285	967	30
397	967	41	321	967	33
453	967	47	228	967	24
465	967	48	604	967	63
299	967	31	227	967	34
214	967	22			

Table 2.
Real Sample Efficiency.

Position	CPM
0	243
90	343
180	293
270	211

Table 3.
Position Effects on Count Rate.

Filter Type	Spike DPM	DPM Found	Percent Efficiency
Clean	1641	1676	102
Clean	1641	1688	103
Clean	1641	1632	100
Clean	1641	1654	101
Dirty	1641	1563	95
Dirty	1641	1567	96
Dirty	1641	1446	88
Dirty	1641	1408	86

Table 4.
Affects of Filter Paper on Alpha Efficiency.

A preparation routine was developed to improve sample recovery. The samples were ashed at 500°C for five hours in the scintillation vials. One milliliter of concentrated nitric acid was added to each vial, and the solution was evaporated to dryness at 200°C. One milliliter of water and 20 mL of scintillator were added to each vial, and the vials were shaken for five minutes. A clear solution was obtained and counted for five minutes. The results of counting seven aliquots of nine different standards are shown in Table 5.

A spiking study was performed on 100 randomly selected air filter cards to determine the effect of the sample matrix on the efficiency of the alpha scintillation system. The filters were cut from the cards, ashed, digested with nitric acid, dissolved in 1 mL of water, and mixed with 20 µL of scintillator. Each sample was counted for five minutes. The samples were removed from the counter, each sample was spiked with 1641 DPM (25 µl) of enriched uranium alpha directly into the cocktail, and another five minute count was obtained. Ten blank filters and ten 1641 DPM spiked, clean filters were counted along with the samples. The sample spike recoveries averaged 92 $\pm$ 5% (SD). Sixteen of the samples had recoveries below 85%. After a visual inspection, 11 of the samples had a faint yellow color and a black residue in the bottom of each vial; three samples had a yellow color only; and two samples had no discernible color. The beta counts for these 16 samples were also higher than the other samples. It was theorized that color quenching had shifted some of the alpha counts into the beta channels. To test this theory, the residual alphas in the beta channel were calculated and the recoveries recalculated. The new recoveries averaged 92 $\pm$ 4% (SD), which agrees with the 92% average of the other 84 samples. Although not solid proof, the data do lend positive support to the theory. The black residue in the 11 samples was analyzed by electron microprobe analysis and was found to be a mixture of Al, Si, Zr, K, Sb, Ti, and Fe. The Fe is thought to be responsible for the yellow color. Table 6 summarizes the results.

All of the spike recoveries were plotted against the transformed Spectral Index of External Standard (tSIE) feature of the instrument to determine if tSIE could be used for quench correction. The feature uses an external ^{133}Ba source (20 µCi) to calculate the tSIE factor (the plot is shown in Figure 2). Apparently, the factor could be used for correction, but further work is necessary to confirm this possibility.

Standard DPM	Average Observed DPM	Average Efficiency (RSD)
0	1.6 +/- 0.3	----
32	29.0 +/- 2.0	90 +/- 9%
63	59.0 +/- 6.0	94 +/- 9%
126	118.0 +/- 6.0	94 +/- 5%
319	295.0 +/- 8.0	92 +/- 3%
623	583.0 +/- 53.0	94 +/- 9%
967	887.0 +/- 28.0	92 +/- 3%
1297	1193.0 +/- 21.0	92 +/- 2%
1641	1479.0 +/- 47.0	90 +/- 3%
		Average: 92 +/- 2%

Table 5.
Alpha Efficiency After Sample Preparation.

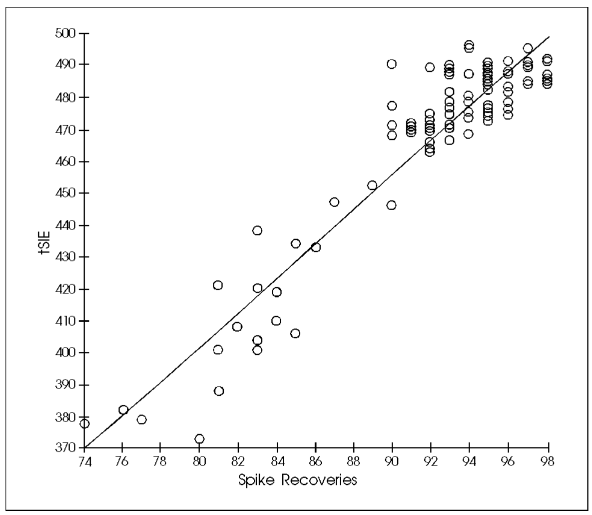

Figure 2.
Spike Recoveries vs. tSIE.

Sample No.	Beta CPM Before Spike	Beta CPM After Spike	Average Beta CPM for Spike	Residual Alpha CPM Misc. Classif. as Beta	Recovery Old (%)	Recovery New (%)
21	2055	2141	284	76	86	91
22	8064	9087	284	739	81	126*
23	4511	4945	284	150	82	91
24	3535	3911	284	92	85	91
25	2174	2549	284	91	84	89
26	3408	3801	284	110	83	90
27	3601	4246	284	362	76	98
28	3466	4191	284	442	77	104
29	2410	2879	284	185	83	95
31	1501	1942	284	157	83	93
32	2010	2351	284	58	84	88
37	543	1079	284	252	74	90
38	306	692	284	103	81	88
91	514	901	284	104	83	90
92	683	1092	284	126	81	89
94	568	1079	284	228	80	94
				New average = 92 +/- 4% (SD)		

Table 6.
Sample Recoveries Corrected for Color Quenching.

Ninety-three air filter samples were randomly selected for a comparison study between the automated air counters, the interim proportional counting method, and the Pulse Decay Analysis (PDA) scintillation counter. Each filter was counted on an in-house fabricated, automated gross alpha counter for one minute. The filters were then cut out of the cards, ashed at 500°C for five hours, and evaporated to dryness with 1 mL of concentrated nitric acid. Two mL of 4<u>M</u> nitric acid were added to each vial. One milliliter was pipetted onto a two-inch stainless steel planchet and evaporated. The planchets were then counted in windowless gas flow proportional counters for ten minutes. The solution left in the vial was evaporated to dryness. One milliliter of water and 20 mL of scintillator were added to each vial, and the vials were shaken for five minutes. Each sample was then counted for five minutes in the scintillation counter.

Ten blank filters and ten filters spiked with 967 DPM of enriched uranium alpha were prepared and counted in both the proportional and scintillation counters. The blanks counted in the proportional counter averaged 0.78 ± 0.45 DPM (SD). Spike recoveries averaged 96 ± 3% (SD). The scintillation blanks averaged 3.3 ± 0.9 (SD) CPM, with the recoveries averaging 93 ± 3% (SD). The comparison data, which have been corrected for the above recoveries, are plotted in Figure 3. As shown in the graph, all three methods agree quite well.

It is of interest to note that MDA's for the liquid scintillation method are as low as that achieved with the gas flow proportional counting method despite the fact that the samples were counted for five minutes with the LSC method as opposed to ten minutes by the gas flow method. The LSC MDA for alpha with a five minute count time is approximately 0.11 pCi/m^3 or 0.24 DPM/m^3. The MDA for alpha using the gas flow method with a ten minute count time is approximately 0.07 pCi/m^3 or 0.15 DPM/m^3. These values are well below the Plant Action Value imposed limit of 4.4 DPM/m^3.

In conclusion, the alpha liquid scintillation counter has the following advantages: (1) high alpha efficiency (>90%); (2) reasonably low background counts (3 to 4 CPM); (3) able to count large numbers of samples automatically; and (4) automatic quench correction. Work continues on experimenting with equipping the instrument with an on-line bar code reader and with possible discrimination of radon daughters.

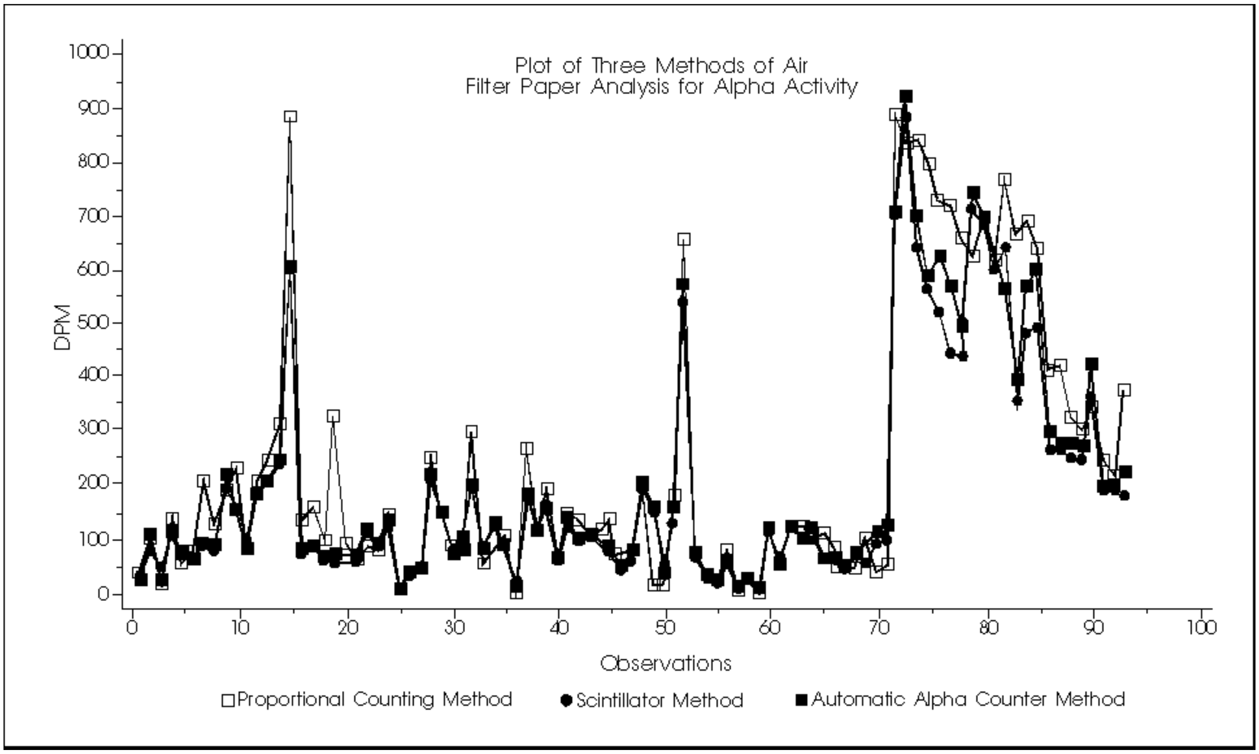

Figure 3.
Comparison of Automatic, Interim, and Scintillation Methods.