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THE EFFECT OF RTP (RADON THORON PROGENY) AND DUST LOADING ON THE DESIGN OF AN ALARM SYSTEM FOR AIRBORNE PLUTONIUM PARTICULATES

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ABSTRACT

If the alpha method is adopted, the main problem for determining airborne plutonium particulates is the overlapping of the alpha spectrum between Pu and RTP (radon thoron progeny). The order of magnitude estimation establishes that RTP is more than 20 times higher than Pu. Therefore a method of discriminating RTP is required. The primary methods of discrimination are the aerosol size and the alpha spectrum methods. If the alpha spectrum method is adopted, the formation of the low energy tail of RTP should be investigated. Preliminary study indicates that the low energy tail is related to the air gap and dust loading.

Both the alpha and thermal neutron-induced fission activation methods can be used to measure the plutonium (Pu) particulates sampled on the filter paper. If the alpha method is used, the primary background is originated from the overlapping of the alpha energy spectra of Pu-239 (E = 5.15 MeV) and the 6 MeV group (Po-218 or RaA and Bi-212 or ThC) of RTP. The overlapping can be well identified in the following alpha spectrum (Figure 1) taken at a uranium enrichment plant[1].

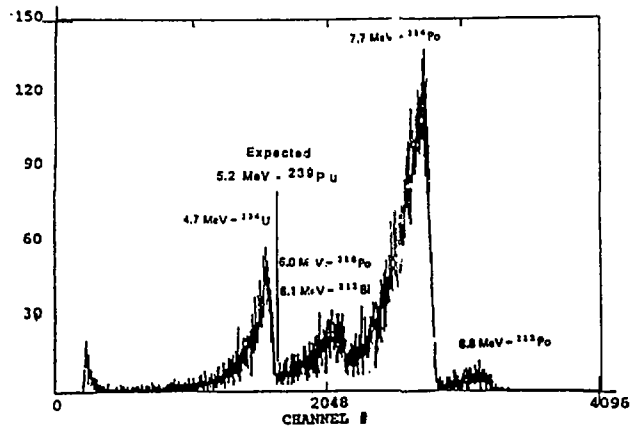

Figure 1

In order to estimate the yields of Pu and RaA accumulated on the filter paper, we use the well-known air sampling formula,

$$I = QC\tau[1 - \exp(-\frac{t}{\tau})] \qquad (1)$$

where I is the number of pCi accumulated on the filter paper in time t, Q is the pump flowrate in litres/minute, C is the concentration of the alpha emitter in air in pCi, and τ is the mean life of the alpha emitter in minutes.

For RaA, $I \simeq QC\tau$ (2)

$t >> \tau$ (4.4 minutes)

For Pu-239, $I \simeq QCt$ (3)

$t << \tau$ (1.82×10^9 minutes)

Then the yield in dpm can be estimated in Table 1.

Table 1

Alpha Emitter	DAC 1 DAC.h	1 DAC.h	Remarks
Pu	60	7.5	(2 x 10^{-3} pCi/L)(flowrate 28 L/min.) (60 min.)(hours of sampling) (2.22 dpm/pCi)
RaA (room)	205	205	(1.5 pCi/L)(28 L/min.)(equilibrium factor, 50%)(lifetime of RaA, 4.4 min.)(2.22 dpm/pCi)
RaA (field)	55	55	(4.2 pCi/L)(28 L/min.)(4.4 min.) (2.22 dpm/pCi)

For Pu, the Derived Air Concentration (DAC) for W-class aerosols is 2×10^{-3} pCi/L (74 mBq/m^3) and alarm should be enabled when the Pu concentration is over 1 DAC according to the United States Department of Energy. The average radon concentration in the United States is around 1.5 pCi/L indoor and 0.2 pCi/L outdoor. The yield of RaA either indoor or outdoor is a constant and is independent of the time of sampling as long as the sampling time is much higher

than 4.4 minutes. The yield of Pu is directly proportional to the sampling time. Unfortunately, for alarm purpose, the sampling time cannot be too long. For one hour sampling, the dpm of RaA is more than 27 times higher than that of Pu. Assuming that 20% of the alpha spectrum of RaA will overlap with the alpha spectrum of Pu, alphas from RaA in the region of interest of Pu are over 5 times more than those from Pu. Therefore a method of discriminating RaA is necessary.

RaA can be discriminated by the waiting time method. That is, Pu can be detected without any interference by waiting for the total decay of the 3 minutes half life of RaA, or around 20 minutes (7 half lives). The waiting time method is absolutely effective, no matter how high the level of RaA is.

There is another component, ThC of the 6 MeV group according to Figure 1. Although the alpha spectra of RaA and ThC are indistinquishable, the amount of ThC can be estimated as half of the yield of Po-212 or ThC' according to their branching ratio. The waiting time method is inapplicable for the discrimination of ThC because it requires about 7 days for its decay. There are two methods commonly used for the discrimination, the aerosols size method and the alpha spectrum method. Both methods are not absolute. That is, the discriminating efficiency depends upon the amount of RTP and dust loading in air.

The aerosols size method was initially reported by CEA (Commissariat l'Energie Atomique) at the 1985 DOE Workplace Workshop. The aerosols first passed through a specially designed channel where the unattached RTP was trapped. The remaining aerosols were sampled through a 3 μM pore size Nuclepore membrane filter paper with cut-off diameter of 1 μM particulates, where the RTP attached to the 0.1 - 0.2 μM aerosols could be effectively removed. Although some of the currently available instruments adopt the aerosols size method, further improvement is still necessary.

The spectrum unfolding method is commonly used in gamma spectroscopy for the separation of two overlapping gamma peaks. However, the unfolding technique for the gamma spectrum is not directly applicable to the alpha spectrum because the latter is characterized by the "low energy tail" which does not exist in the gamma spectrum. An effective alpha spectrum method should base upon an understanding of the low energy tail. The following two figures will help to show the nature of the low energy tail of

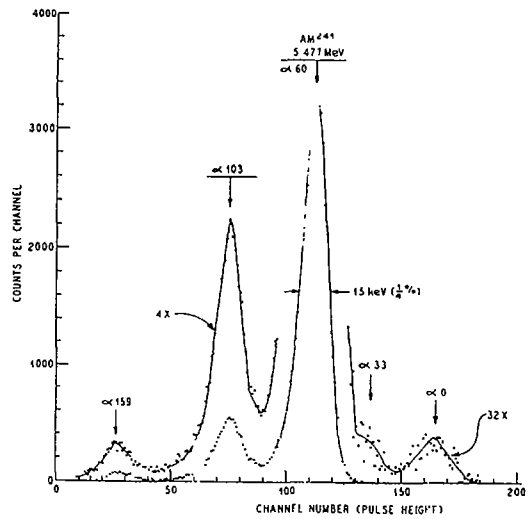

Fig. 14 Alpha-particle spectrum of Am^{241} measured with a gold-silicon surface barrier detector (ref. 67)

Figure 2

the alpha spectrum. Figure 2 shows an alpha spectrum for Am-241 using a thin electroplated source measured under vacuum[2]. It can be seen that the alpha spectrum is very close to symmetric and the low energy tail is nearly non-existing. Figure 3 shows the alpha spectrum of U-234 particulates of size 1.0 μM collected on a

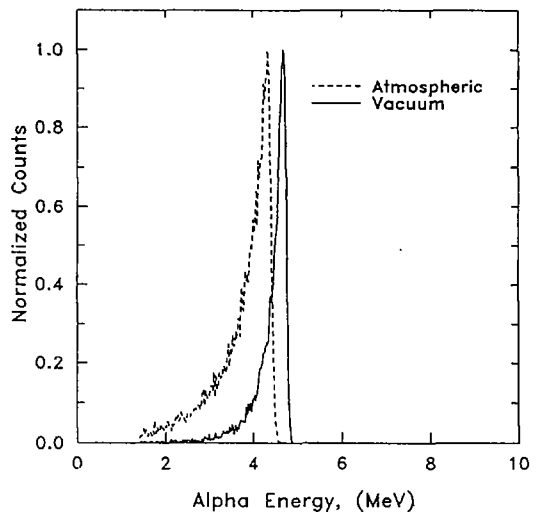

Fig. 1. Spectra from 1.0-μm diameter uranium (^{234}U) acetate particles deposited on 1.2-μm pore size Millipore filters. Analyses performed at atmospheric pressure and under vacuum. Source-to-detector gap = 5.0 mm.

Figure 3[3]

filter paper of 1.2 μM pore size. The peak energy of the 4.7 MeV alphas (under vacuum) after passaging through an air gap of 5 mm. will be degraded to 4.2±0.2 MeV according to the range energy curve. This explains the shift of the peak value between the spectra measured in the atmosphere and under vacuum respectively in Figure 3. The spread of the 0.2 MeV is estimated based upon the intercept of the high energy side of the spectrum on the energy-axis. Assuming that the extension of the "low energy tail" is due to the energy degradation of the 4.0 MeV alphas (the intercept of the low energy side is assumed to be symetric) after passaging along an oblique direction rather than perpendicular, the oblique distance corresponding to 75° is 5 mm./cos 75° = 19 mm. Again, according to the range energy curve, the range of the ~4 MeV alphas in air is 26 mm. After passaging through an air gap of 19 mm., the ~4 MeV alphas will be degraded to ~1 MeV which is consistent with the end-point of the alpha spectrum in the atmosphere in Figure 3. The less extended low energy tail of the alpha spectrum of 4.7 MeV alphas measured under vacuum can be explained by the partial burial of the uranium particulates in the filter paper.

In analog to the formation of the low energy tail in air, the effect of dust loading on the low energy tail can be approximately predicted by preparing a range energy curve based upon the well-established NASA SP-3013, Berkas & Berger, "Table of Energy Losses and Range of Heavy Charged Particles", 1964. Figure 4 is an example of a range energy curve for both ordinary soil of aluminum silicate compound of polycarbonates (Curve I).

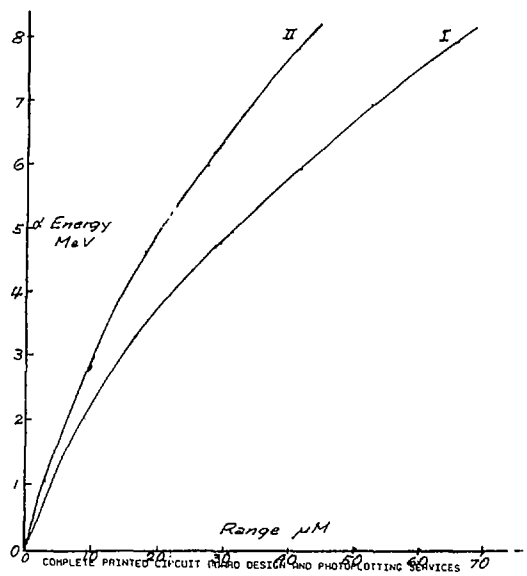

Figure 4

Table 2 gives the estimation of the deposition thickness in μM for filter paper (detector) of different sizes. Since it only needs 4 μM of the ordinary dust to degrade alphas of 6 MeV to 5.4 MeV according to Figure 4, the critical angle can then be calculated, according to Figure 5. Here 5.4 MeV is the upper limit

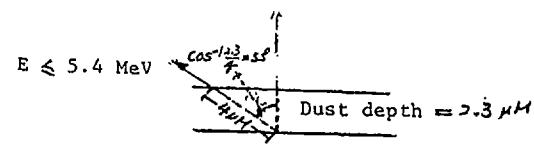

Figure 5

energy on the high energy side of the alpha spectrum of Pu-239. According to Table 2,

Table 2

Size (cm ID)	Area (cm^2) (Actual area for collection of dust)	mg/cm^2	Thickness in μM mg/cm^2/2680 mg/cm^3 (ordinary silicon dust)	Critical angle θ_c
2.5	$\pi(1.1)^2$ = 3.8	8.4/3.8 = 2.2	2.2/2680 = 8	0°
4.7	$\pi(2.1)^2$ = 13.8	8.4/13.8 = 0.61	0.61/2680 = 2.3	55°
6.0	$\pi(2.8)^2$ = 24.6	8.4/24.6 = 0.34	0.34/2680 = 1.3	71°
8.0	$\pi(3.8)^2$ = 45.4	8.4/45.4 = 0.19	0.19/2680 = 0.7	80°
10.0 (4")	$\pi(4.5)^2$ = 63.6	8.4/63.6 = 0.13	0.13/2680 = 0.5	83°
20.0	$\pi(9.8)^2$ = 301.7	8.4/301.7 = 0.028	0.028/2680 = 0.1	88°

discrimination of the alphas from RTP can either be performed by increasing the size of the filter paper (detector) or by limiting the incident angle by a collimator. Instruments currently available using collimators were designed for air gap correction only, they cannot be used for dust loading. On the other hand, gas proportional counters of large area (8 to 20 cm ID) are available so that the size method is favoured. If the size method is adopted, the minimum size of the filter paper (detector) is 4.7 cm ID. Experimental verification of the above can be conducted using a radon box.

Finally, there are three remarks which should be mentioned. Firstly, both CAM (continuous air monitor) and the sequential device can be available for the alpha method. The sequential device uses an open-face air sampler and a separately located counter-scaler. CAM is essentially required when the lifetime of the nuclides measured is shorter than the instrument

integrating time, for instance, the measurement of short-lived RTP. However, for the measurement of Pu alphas with longer lifetime, it is not entirely necessary. The sequential device is more flexible in the choice of counters of different types and sizes, and in the choice of different discrimination methods. On the other hand, both the choice of detectors and the discrimination method are limited for CAM.

Secondly, prompt on-site discrimination against RTP is only required for the alarm system. For area monitors which are used for providing records of Pu levels and personal doses in the plutonium workplace, prompt on-site discrimination is not required because we can always wait for the complete decay of RTP.

Thirdly, as indicated at the beginning of this work, the thermal neutron-induced fission method will not be interfered by the existence of RTP and can be adopted whenever the alpha method is inadequate.

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Electrodeposition of Selected Alpha-Emitting Nuclides from Ammonium Acetate Electrolyte

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