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Direct-Scanning Alpha Spectrometer for Americium and Plutonium Contamination on Highly-Enriched Uranium Surfaces

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Abstract

Trace Pu^{239} and Am^{241} contamination on a surface whose alpha count is dominated by U^{235} and U^{234} decay has been successfully quantified by counting swipes in external alpha spectroscopy chambers. The swipe process, however, is labor intensive and subject to uncertainties in the swiping process as well as degraded spectral resolution due to the presence of the swipe material. A multichannel instrument for automated in situ measurements of interior and exterior contamination has been developed which incorporates a rotary table, 13 fixed ion-implanted silicon detectors, and spectroscopy electronics. Custom software was written to allow alpha spectrometer to function as a virtual instrument in the LabView environment. This system gives improved speed and resolution as well as a complete log of the location of areas of high surface contamination, a feature not practical to obtain by other methods, and one which opens the possibility of long term studies such as Pu outgrowth evaluation employing the instrument. We present performance data as well as system integration, calibration, control, and dynamic geometric efficiency calculations related to the design of this and next generation systems.

Introduction

The push for stockpile reduction has produced a significant amount of uranium which must be decontaminated¹ and measured for safe storage or further processing. Americium and plutonium contaminants must be verified to average less than 20 dpm/100 cm² swipable and below 500 dpm/100 cm² direct surface contamination. While instruments and procedures for measuring swipable contamination are well established, the hardware and analysis required for the direct measurements has yet to be produced and verified; this paper reports progress on this effort.

The direct-counting instrument design presents several challenges. While there is sufficient separation between the highest uranium alpha energy (4.768 MeV) and the various Pu and Am peaks (5.096–5.482 MeV), the amplitude of the primary uranium spectrum can be more the 3 orders of magnitude greater than the maximum permissible contamination level. This level of decay is well above what the chosen instruments can resolve without some form of physical barrier to reduce the count rate, and further, the useful lifetime of the detectors is too limited at the full rate of alpha bombardment. The detectors are vulnerable to contamination by recoil, but in this application The instrument complexity is also nontrivial, as the hemispherical parts must be

rotated past the detectors and multiple detectors, carefully arranged, must be scanned to cover the entire surface area. In this configuration, the geometric efficiency of the detectors is also a complex function of position.

The complexity of the direct counting instrument can, however, be exploited to create additional features. Its repeatability is inherently better than for swipe counting, but more significantly, it can produce surface maps of contamination intensity. This information can be used to evaluate cleaning methods or simply to identify areas for additional spot cleaning.

Experimental

The direct alpha spectrometer unit is constructed as a hinged vacuum chamber with 13 surface barrier alpha detectors² arranged in the walls to view both sides of the part in a continuous distribution. The detectors are 300- and 1200mm² in surface area, with the smaller detectors positioned near the top (pole) of the unit. The base of the chamber has a stepper motor-driven turntable ring which rotates the part past the fixed detectors. The detector amplifiers are fed to a 16-input, 1024-channel multichannel buffer (MCB) which is controlled through a memory-mapped PC interface. Figure 1 is a

Figure 1: Alpha spectrometer fixture.

photo of the assembled instrument.

Three components comprise the instrument's software. A preprocessing program calculates geometric efficiency for a shell size and sets detector weights to maximally smooth efficiency averaged over the surface. The control, measurement, and display software, written in the LabView³ environment, links the valves, pressure transducers, turntable, and MCB. It finds the overall contamination level and performs several intermediate calcula-

Figure 2: Typical display panel.

tions before generating output files. Figure 2 gives a typical screen display from the instrument. Post-processing routines manipulate the data and display it as polar plots or 3-D surface maps.

The control program handles gentle evacuation and repressurization of the chamber, background measurement/subtraction and calibration using automatic peak fitting to dual isotope check sources. Although there has been concern that recoil contamination will create rising background levels,

all detectors have so far remained clean. Furthermore, contamination contribution to background at energies in the uranium spectrum, where most of it would be expected to occur, will not affect the instrument's ability to make the intended Am and Pu measurements.

During counting, the shell is rotated in discrete steps, stopping every few degrees. The spectrum is divided into uranium and contaminant components, and a $13 \times N$ array is thus generated for each. Detector weighting can be applied up front, by programming different live times for each detector, or after a count is complete with uniform live time for each detector. The latter approach is preferable when a fine rotational step size is used, resulting in short live times at each station.

Two techniques are being explored for dividing the output spectrum into uranium and contaminant components. There is enough separation between spectra that simple integration of the spectra between fixed energy levels appears to be adequate. This method can be used even when contaminant counts at each rotational position are small (almost always the case when the rotation is performed in fine increments). In this mode, the instrument functions like a flexible, intelligent, multi-input discriminator. The more sophisticated technique is to perform curve fitting⁴ to remove the uranium

Figure 3: Peak fitting to synthesized data comparable to DU measurement.

spectrum. This allows the tail end of the contaminant spectra to be counted more accurately, but requires a longer count time at each position, and is difficult in any case for very high rate uranium spectra. Figure 3 is an illustration of the double peak fitting algorithm operating for simulated contamination on a depleted uranium (DU) part.

To measure highly enriched uranium (HEU) shells, the alpha particle rate must be reduced by a factor of 10 or more to prevent detector or amplifier saturation and generate false excess contamination readings. This also extends the life of the detectors, which would otherwise reach their damage threshold after less than 200 hours. Unfortunately, the minimum

counting time must also be increased.

The *spatial* geometric efficiency of the direct-scanning instrument, that is, its efficiency at counting dpm/cm² due to spot contamination, requires careful consideration. The detectors are arranged with slightly overlapping views from the pole to the equator of both the interior and exterior of the fixture. A detector near the pole is exposed to the same small region of the test specimen for much of the measurement, whereas a detector near the equator sees a portion of the shell only briefly as it rotates in and out of its horizons. Many segments of the specimen are also partially visible to more than one detector. The determination of an average contamination for the whole part requires that these interactions be calculated so that the output of each detector can be weighted in order to generate a uniform spatial geometric efficiency. The efficiency is also sensitive to the radius of the test shell and must be recalculated for each size that the instrument is to measure.

A ray-tracing program was written to calculate the point-to-point geometric efficiency across the shell. Typical calculations for instantaneous efficiencies are depicted in Figure 4, which shows a hot spot under each detector. (Each detector is illustrated by a hollow ring above the surface.) Averaged over a complete part rotation, the efficiency surface takes the shape

Figure 4: Stationary geometric efficiency. The right hand image is a reflection from a 'mirror' plane, displaying the interior of the shell.

of rings centered under each detector. The unweighted variation in this 'ring efficiency' is shown in cross section in Figure 5a. When the output of each individual detector is adjusted, the ring efficiencies can be made much more uniform. The optimal set of weights can only be found by iterative solution, however, and the results are dependent on the shell diameter. In Figure 5b, the cross-section shows the variation in ring efficiencies after a calculated set of weights (with a maximum weight of 1).

Figure 5: Uncorrected (a) and smoothed (b) ring efficiencies.

Results

Two different test shells were evaluated during shakedown of the instrument. The first, a DU shell, required no physical barriers to reduce the overall alpha rate. For the HEU tests, plastic covers were fitted to reduce the effective surface area by 90%. Both shells were clean, and expected to have only trace contamination. On the DU shell, a Pu^{239} check source (2000 dpm) was attached near detector 4 at $\theta = 0$. The spike caused by this source is clearly visible in the left display of Figure 4. Detector 12, which monitors the edge of the edge of the shell, shows much more ambiguous spectra and potentially indicates larger contamination. Some of the surface that the detector sees here is perpendicular its surface, which explains some of the

Figure 6: Two measured contamination slices with representative spectra for the DU test shell. Left (detector 4) has a 2000 dpm check source mounted at $\theta = 0$. Right (detector 12) is monitoring the perpendicular bottom edge.

spectral degradation. It is not yet known whether the increase is due to measurement error or is some product of the bisection or cleaning process.

For the HEU shell, the instrument behaved well at the small (300-mm²) detectors with 90% of its area masked off (Figure 7, detector 9). The larger surface area of the 1200-mm² detector with a similar mask was apparently too much for the amplification and pulse-shaping electronics, however. Pile-up due to coincident arrivals was higher than individual channel counts, and

Figure 7: Screen data for the HEU shell. Left (detector 9) shows the instrument working correctly. Right (detector 12) shows the effects of excessive rate, indicated by high pile-up count.

spectral degradation occurred (Figure 7, detector 12). This effect was intermittent; at some points, the measured contaminations go down to expected levels, and at others (the peak around 35 degrees), the level is even higher.

Conclusion

Although some further adjustments are required, this instrument fulfills its purpose for the evaluation of nuclear material to be removed from the stockpile. It gives useful 3-D information about contamination distribution and

is able to produce rapid estimates (less than 20 minutes) in most cases. The HEU measurement indicate, however, that a higher percentage of the alpha particles must be blocked from entering the detectors for these shells to prevent spectral degradation. There also appears to be a broad and active spectrum coming from the equatorial edge of the shell that still must be understood.

For a next-generation device, it appears that a slower, less expensive, but more flexible device should be developed which scans both the latitude and longitude of the shell with only one or two detectors devoted to each surface. This would allow for simpler electronics, at the expense of slightly more elaborate hardware. The elaborate process of calculating geometric efficiency would also be simplified, and calibration would be dramatically easier. Multiple units of this type might be built if the speed of a single unit is not adequate.

Further development will be done to evaluate whether thin alpha absorbers on each detector will allow better discrimination in the resulting spectrum. The increase in spectral dispersion will be traded off against the benefit of reducing the count from the lower energy particles to a more reasonable level.

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