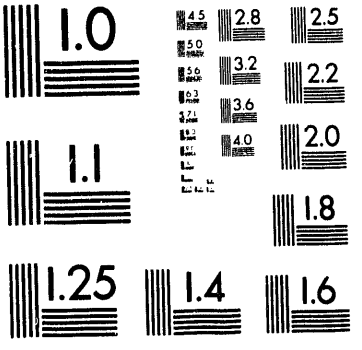
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TITLE: *TEST RESULTS OF A NEW DETECTOR SYSTEM FOR GAMMA RAY ISOTOPIC MEASUREMENTS*

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TEST RESULTS OF A NEW DETECTOR SYSTEM FOR GAMMA-RAY ISOTOPIC MEASUREMENTS

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ABSTRACT

A new type of gamma-ray detector system for isotopic measurements has been developed. This new system, a "Duo detector" array, consists of two intrinsic germanium detectors, a planar followed by a coaxial mounted on the same axis within a single cryostat assembly. This configuration allows the isotopic analysis system to take advantage of spectral data results that are collected simultaneously from different gamma-ray energy regimes. Princeton Gamma Tech (PGT) produced several prototypes of this Duo detector array which were then tested by Rocky Flats personnel until the design was optimized. An application for this detector design is in automated, roboticized NDA systems such as those being developed at the Los Alamos TA-55 Plutonium Facility. The Duo detector design reduces the space necessary for the isotopic instrument by a factor of two (only one liquid nitrogen dewar is needed), and also reduces the complexity of the mechanical systems and controlling software. Data will be presented on measurements of nuclear material with a Duo detector for a wide variety of matrices. Results indicate that the maximum count rate can be increased up to 100,000 counts per second yet maintaining excellent resolution and energy rate product.

INTRODUCTION

Within the past decade, great strides have been made in gamma-ray spectrometer systems for plutonium isotopic measurements. It has been shown[1] that it is possible to achieve very high count rates approaching 70 kHz with throughput rates (counts stored per unit real time) exceeding 30 kHz while maintaining good energy resolution (500 to 600 eV at 122 keV). This was 4 to 5 times higher than the traditional counting rates at the time[2-4] and was achieved primarily because of advances in detector/preamplifier technology as well as proper coupling with an amplifier and analog-to-digital converter optimized for high throughput. With an optimized system, precisions for the effective specific power of 1% or better could be achieved in counting times of 10 minutes on a wide variety of plutonium product and scrap forms.

Due to changes in the regulatory environment, priorities have shifted from production to dismantlement [5-7] with increased emphasis on ES&H (environment, safety, and health) and quality assuance related issues. These issues are partly responsible for the drive toward utilizing expert systems including robotics and artificial intelligence. Several years ago an automated calorimetry and gamma-ray isotopics system (ROBOCAL [8,9]) was installed at the Plutonium Facility at Los Alamos National Laboratory. This is a full-scale, prototypical robotic system for remote calorimetric and gamma-ray analysis of special nuclear materials. Many robotic systems of the gantry type, which includes ROBOCAL, work within a confined area typically referred to as a "work envelope." It therefore becomes very important to utilize the available resources, most notably, space, with maximum efficiency.

A new detector system which reduces the space requirement (footprint) by a factor of two while maintaining the full range of detection capability (both low and high energy regimes as well as high count rate) has been developed at the Rocky Flats Plant in conjunction with the detector manufacturer Princeton Gamma Tech (PGT). An additional advantage realized by this new system is a reduction in software complexity when applied to systems such as ROBOCAL since only one detector platform and absorber/collimator assembly needs to be controlled.

DEVELOPMENT OF DUO DETECTOR SYSTEM

Historical Progression

The advantages for improving the precision and accuracy of Pu isotopic measurement work by utilizing two energy regimes (100-450 keV and 600-1000 keV) have been demonstrated earlier [2]. The basic finding of this early study was that isotopic information from the high energy region could be combined with the low energy results for improved precision in the measured isotopic ratios. In addition, certain isotopic compositions

benefitted from improved accuracy by utilizing the high energy regime; e.g., low Am concentration materials.

The spectral information from the high energy coaxial detector regime is also useful for other NDA measurement situations. Quite often SNM product, scrap, or waste forms are packaged in thick walled containers that may or may not have lead lining for personnel dose reduction. In these cases, the lower energy gamma rays (100-450 keV) may become severely attenuated and the high energy gamma rays provide much improved accuracy and precision over the low energy results.

Other applications of the two detector method which have become much more important recently are: measurements for dismantlement verifications or for anti-terrorist operations; situations where large amounts of absorber can be between the SNM and the gamma-ray detector. In these cases, it is not unusual for the gamma rays below 300 keV to be completely obliterated. The spectral information from the more penetrating gamma rays in the coaxial detector, however, can still be utilized to obtain the requisite isotopic information for various measurement applications. (One of the options of the TRIFID [10] analysis software allows for just this type of isotopic analysis where the low energy gamma rays are almost totally absorbed before reaching the detector.)

Isotopic analysis systems, e.g., TRIFID, that employ dual range spectral measurement capabilities utilize two separate germanium detectors, a high resolution planar and a high efficiency coaxial, with the attendant space requirement for two liquid nitrogen dewars. As stated above most laboratory, portable, and roboticized NDA measurement applications could benefit from a reduction in the instrument footprint that could be realized by incorporating both detectors into one package.

In 1986, in connection with the isotopic high count rate development work at Rocky Flats[1], several detector manufacturers were contacted about the feasibility of building a detector with two dissimilar crystals (planar and coaxial) in one cryostat; the "Duo" detector arrangement. Technical design specifications for the detector crystals were kept identical to that for the previous individual detector packages, but for the Duo detector arrangement the crystals were to be contained in one cryostat, the coaxial detector positioned behind the planar on the same axis. One manufacturer, Princeton Gamma Tech, began the development of a prototype detector to meet these requirements.

The first prototype detector was delivered and evaluated at Rocky Flats in mid 1988. This first prototype detector and later a second in 1989 did not meet all the performance specifications and were returned to the vendor for rework. The first Duo detector that met (and exceeded) the original performance specifications was received at Rocky Flats in May 1991.

Duo Detector Design

The Duo detector design is an array of two germanium crystals mounted on the same axis within an eight inch long aluminum endcap. The crystal closest to the face of the endcap is an intrinsic germanium planar detector mounted within 5 mm of the endcap. The coaxial detector is mounted behind the planar on the same axis and is constructed of n-type material for neutron damage resistance. A lead absorber, 2.5 mm in thickness, is positioned between the two crystals to attenuate the low energy photons entering the coaxial detector (these events are collected by the planar detector for analysis and counting these in the coaxial would be redundant). No other absorbers are utilized since the high energy gamma rays of interest are typically already of low intrinsic intensity, and also since the energy rate product for the coaxial detector is typically much higher than that for a planar and therefore can manage much higher count rates. A schematic of the crystal arrangement is shown in Figure 1.

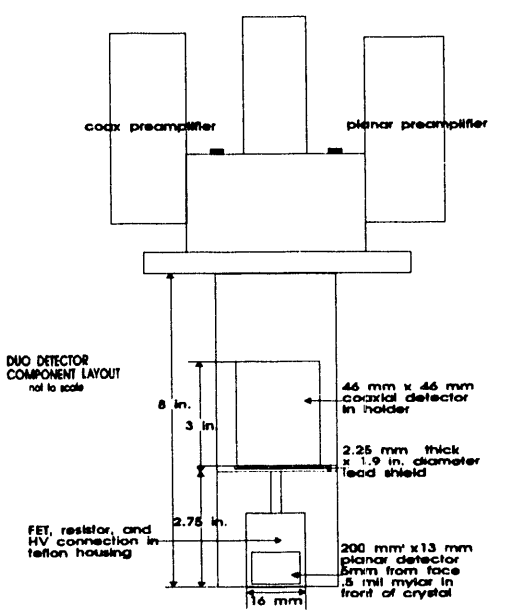

Figure 1. Schematic representation of a cross-sectional view of Duo detector cryostat assembly.

The planar detector has a surface area of 200 mm ^{2} and is 13 mm in depth. This size detector was chosen as a tradeoff between reasonable efficiency and extremely good resolution. The original design specifications specified a resolution of less than 580 eV at 122 keV for a 1 micro-second amplifier shaping time constant at 50,000 counts per second. Peak shape criteria were

specified as the full-width-at-tenth-maximum to full-width-at-half-maximum ratio (FWTM/FWHM) to be less than 1.9 and the full-width-at-fiftieth-maximum to full-width-at-half-maximum (FWFM/FWHM) to be less than 2.7.

The coaxial detector crystal is typically 45 mm in diameter by 45mm in depth. The relative efficiency for this size detector is approximately 12%. This size detector was chosen as a reasonable tradeoff between high efficiency for the gamma-rays of interest while maintaining good resolution. The original design specifications specified a resolution of less than 1.7 keV at 661 keV for a 1 microsecond amplifier shaping time constant at 50,000 counts per second. Peak shape criteria were identical to those specified for the planar crystal. Both the planar and the coaxial crystals are supplied with separate preamplifiers.

Rocky Flats Plant (RFP): Initial Test Results

After receipt of the first operational Duo detector in May 1991, performance tests were conducted to assess the count rate and resolution performance of the unit. Tests were conducted with sealed point sources with activities as high as the hundred microcurie range to obtain high count rates. Of primary importance for the planar crystal is the resolution as a function of count rate and the total energy rate product (count rate x average energy deposited). Purchase specifications for the resolution were noted above as: less than 580 eV at 122 keV with a 1 microsecond amplifier shaping constant at 50,000 counts per second. This corresponds to an energy rate product of about 6,000 MeV/sec for ^{57}Co. (These performance specifications were fairly typical of the detector industry's manufacturing capability for individual detector units at that time.) The high resolution for the planar is a necessity for Pu isotopic work since the gamma-ray lines of interest are very tightly grouped in the 100-450 keV energy region.

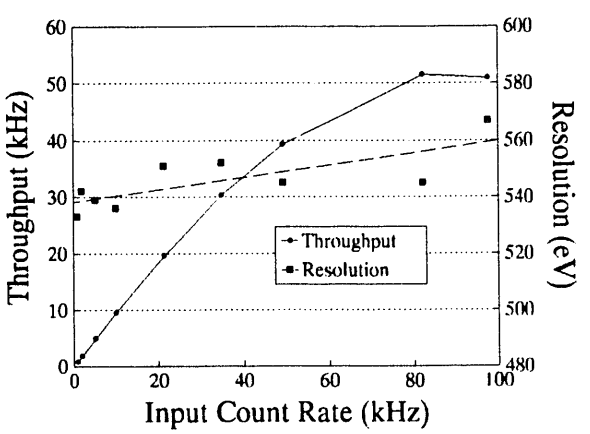

Figure 2. Throughput and resolution as a function of the planar detector input count rate.

Figure 2 shows the count rate and the resolution performance of the first Duo unit planar tested. Incoming count rates as high as 100,000 counts per second were achieved without suffering preamplifier saturation. This count rate corresponds to an energy rate product in excess of 10,000 MeV/sec, clearly exceeding purchase specifications. Resolution at even the highest count rates was below 570 eV, again easily meeting the purchase specifications. Note in Figure 2 that the throughput rate (counts stored per unit real time) is still rising at around 80,000 counts per second input count rate. This extends the conclusions of Reference 1 and illustrates that input count rates approaching 100,000 counts per second can be utilized at 1 microsecond shaping time without any loss of throughput rate.

Resolution is not as important an issue for the coaxial crystal as it is for the planar since the gamma-ray lines tend to be separated better in the high energy region. Initial tests of the coaxial crystal showed that the performance specification of 1.7 keV at 661 keV for 50,000 counts per second could be achieved with a 1 microsecond amplifier time constant. These tests were conducted with a ^{137}Cs point source. Tests were also conducted for higher count rate performance. Count rates in excess of 150,000 counts per second were routinely achieved with acceptable resolution and good spectral quality.

Table I. Initial Duo detector results for Pu oxides and molten salt residues.

Item Type	Crystal Type	Count Rate (kHz)	Resolution (eV)
Oxide 1	Planar	55.7	564
	Coax	49.0	1620
Oxide 2	Planar	55.7	569
	Coax	44.0	1590
Salt 1	Planar	48.0	568
	Coax	174.0	1680
Salt 2	Planar	98.0	561
	Coax	101.0	1600

Following the point source tests, the Duo detector was tested with SNM sources. (Due to the curtailment of activities at Rocky Flats in the early 90's, these tests were by necessity somewhat limited in scope.) The initial testing was performed with oxides and molten salt residue items since these items typically generate the largest absolute count rates. Spectra were obtained on these items with the largest count rates obtainable and the spectra were analyzed using the TRIFID isotopic analysis code. Results

of these tests are listed in Table 1. The listed resolutions were determined by the TRIFID isotopic analysis code and are given for the 129 keV ^{239}Pu and the 662 keV ^{241}Am gamma ray lines for the planar and coaxial crystals respectively. Amplifier shaping constants were set at 1 microsecond for both crystals.

Los Alamos National Laboratory (LANL): Experimental Results

Approximately a year ago, a PGT Duo detector with identical specifications to that already described was purchased by the Los Alamos Nuclear Materials Measurement and Accountability Group for possible application with robotic systems such as ROBOCAL.

The Los Alamos Duo detector is installed in an isotopics detector stand which gives the capability of translation and rotation of the samples as they are assayed. The stand provides lead shielding around the detector endcap as well as the sample enclosure. There is a one to two inch gap at the end of the detector endcap where a lead collimator can be installed for heterogeneous items. The collimator is 1 inch to 2 inches thick with a variable 0.5 to 1 inch wide by 2.5 inch long aperture.

The data collection electronics consist of a Canberra Series 95 multichannel analyzer, two Tennelec TC 244 amplifiers, two Ortec 459 high voltage supplies, an Ortec 974 counter timer, and two Canberra 8076 analog to digital converters. The spectral data were again analyzed using the TRIFID isotopic analysis code.

Measurements were completed on a variety of samples and on standards to evaluate the performance of the detector in a working environment.

Discussion of Results

Since both crystals reside in a single assembly with a fixed geometry in the Duo detector, it was decided to determine the count rate response for both crystals as a function of source distance from the endcap. The measurements were made on a well-characterized oxide standard, both with and without a collimator, and the results are exhibited in Figure 3. Note that the planar count rate typically exceeds that of the coaxial detector. However, for the uncollimated case, a crossover between the count rates occurs at a distance of 6.5 inches.

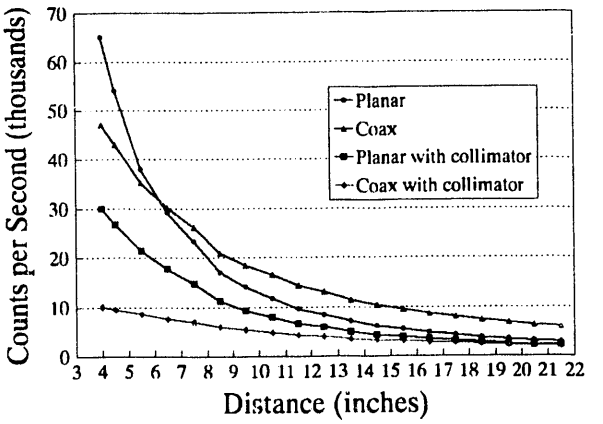

Figure 3. Countrate as a function of distance for the planar and coaxial crystals both with and without collimation.

Then measurements at four different counting times were made on several different SNM material types with varying matrices in order to evaluate the time dependence of the effective specific power precision of the Duo detector measurement results. These items are difficult to assay with a good precision and accuracy because of contaminants, heterogeneities, and self-absorption. The first category contains three varieties of heterogeneous salts with a high americium content. The second measurement category consisted of two oxide types.

The first set of measurement items are salt residues from a molten salt extraction process and the precision for the effective specific power results are shown in Figure 4. These normally vary in heterogeneity and are usually high in americium. The next set of measurement results, shown in Figure 5, are from an electro-refining process. These salts contain crucible parts with the electro-refining salts adhering to the parts. The final set of salt results, shown in Figure 6, are from an oxygen sparging process with most of the plutonium residing in the salts. These are normally very heterogeneous with high levels of americium.

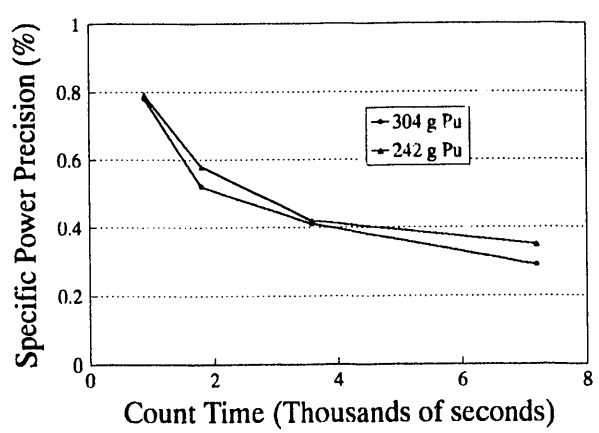

Figure 4. Precision of the effective specific power as a function of counting time for two salt residues from a molten salt extraction process.

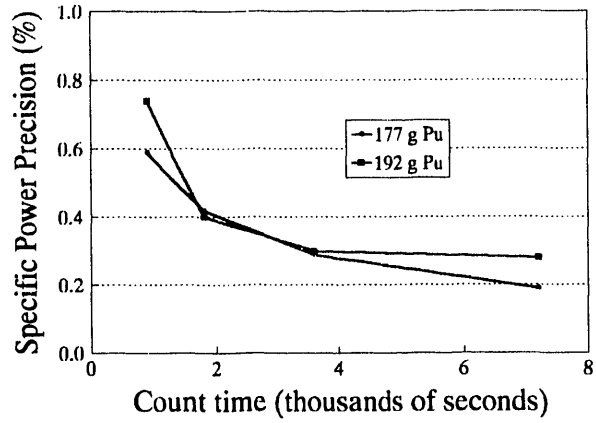

Figure 5 Precision of the effective specific power of counting time for two salt residues from an electro-refining salt.

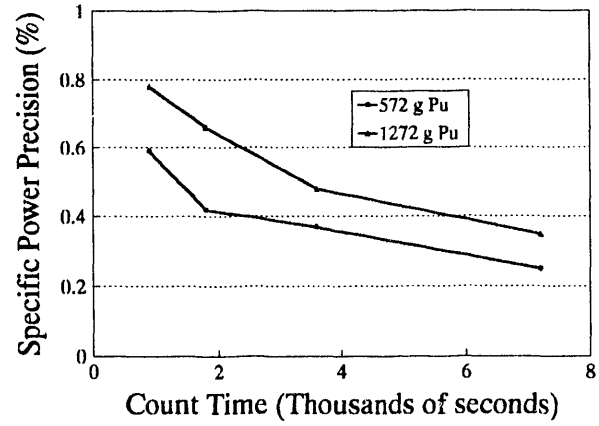

Figure 6. Precision of the effective specific power as a function of counting time for two oxygen sparged salts.

The next measurement group is the oxide category. Measurement results from impure oxides which contain significant levels of tantalum and tungsten are shown in Figure 7. The final set of results, shown in Figure 8, are from pure oxides that are highly enriched (35 to 40%) in the isotope ^{240}Pu. These items are from recent ion-exchange-separated material and are very low in americium (less than 1,000ppm). Here the coaxial detector results are crucial to obtain any reasonable americium information.

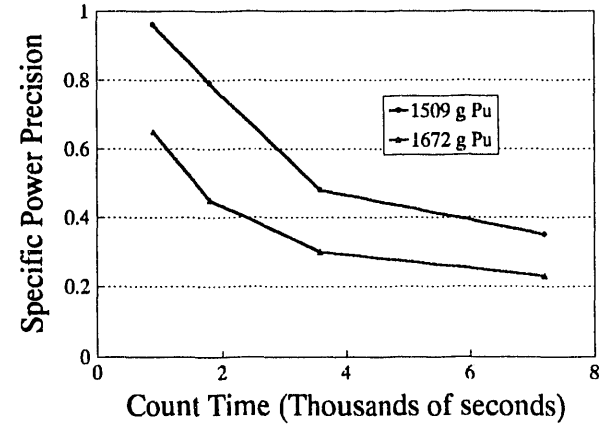

Figure 7. Precisions of the effective specific power as a function of counting time for two oxides significantly contaminated with tantalum and tungsten.

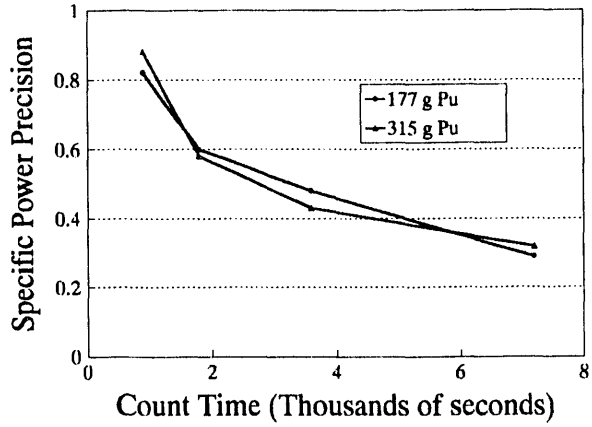

Figure 8. Precision of the effective specific power as a function of counting time for two pure oxides enriched to about 40% in ^{240}Pu and having less than 1000 ppm americium.

The above figures clearly show that the uncertainty of the effective specific power can be obtained to less than 1% for a count length of 15 minutes and in most cases to about 0.5% for 30 minutes. This performance is on the same level as that described in Reference 1 for the two-detector system on similar items. There would almost never be a reason to count longer than 30 minutes, as the data indicates that very little additional precision would be gained.

SUMMARY/CONCLUSION

The Duo detector is advantageous for systems where data from both the low and high gamma-ray energy regimes are needed and where minimal space is available. Since the level of complexity is also reduced, the Duo detector is ideal for automated, robotic systems where space is at a premium and reduced complexity desirable for computer applications. Manual operations are also improved due to having only one detector/dewar configuration to position.

The data presented in this study show that effective specific power precisions of 1% can be attained for count times of less than 15 minutes for the Duo detector. In most cases, precisions of less than 0.5% were reached in less than 30 minutes. This practically eliminates the need to have count times longer than 30 minutes.

In the past, the downside of the two-detector system was the physical requirement of having two detectors. Now, with the Duo detector system, there is a reduction of the overall complexity of the isotopics analysis system and there is no degradation in the quality of the measurement or spectral results.

ACKNOWLEDGEMENTS

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well as information on the items, is greatly appreciated. Sandra Hildner was of invaluable assistance in the retrieving of items and preparing them for measurement. The expert help provided by Sandra Ball on the word processor and on the final formatting of this paper was invaluable. Teresa Cremers' help on the inclusion of the data charts and graphs to the body of this study is greatly appreciated.

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