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DOING PHOTONS WITH MERLIN II AT OROVILLE

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ABSTRACT

A very large n-type high-purity Ge-semiconductor detector has recently been installed in the Lawrence Berkeley Laboratory's underground low-background facility at Oroville. The detector (named MERLIN II) is mounted in a low-activity cryostat and has a rated "efficiency" of about 115% - - nearly 4 times higher than our original MERLIN detector, which was used extensively to measure the minute amounts of radioactivity in samples from the LDEF satellite. We discuss gamma-spectrometric analyses with MERLIN II on some of the same LDEF samples, providing direct evidence for improvement in performance achieved with the larger detector.

INTRODUCTION

Radiometric analysis of materials retrieved from the LDEF satellite has continued at the LBL Low Background Facility from the time of first receipt (March 1990) until the present. Results of our analyses, along with results from many other members of the radiometric analysis group, are summarized in References 1 and 2. Our lowest background (BKG) detector, the MERLIN I detector at Oroville, was used extensively to measure the smallest activities until its failure in Spring 1991. The MERLIN I detector could not be revived. It was replaced in January 1993 by a similar type detector of much larger size (efficiency), for which a major fraction of counting time has been applied to further analysis of LDEF samples. Some important characteristics of the new detector (MERLIN II) are documented here, along with examples of results obtained with the MERLIN II system on LDEF samples.

The MERLIN II Detector

The Lawrence Berkeley Laboratory's Low-Background Facility operates low-level counting installations at two sites: at the LBL Berkeley Site and in the underground power plant of the Oroville Dam (a facility of the California Department of Water Resources). We have recently installed a new gamma-spectrometer detector system at the Oroville Site, where the overhead rock shielding reduces the surface Cosmic Ray intensity by 1000-fold. The new detector provides nearly a 4-fold increase in detection sensitivity compared to our previous capability, while at the same time maintaining the very low BKG characteristics achieved formerly with a much smaller detector. Since the LBL Low Background Facility is a national User's Facility, it is appropriate to make this new capability known to the community of potential users.

The "MERLIN II" detector, a replacement for our "MERLIN I" detector, is a much larger n-type high purity Ge detector than was our original "MERLIN I" detector. "MERLIN II" has a rated "efficiency" of about 115%, while "MERLIN I" was rated at about 30%. The new detector is 80 mm in diameter and 85 mm in length, weighing about 3 Kg. It is mounted in the manufacturer's (Ortec) "low-BKG" cryostat assembly, for which LBL supplied some special low-activity components, including the boron nitride stand-off insulator and the front-end FET package.

The MERLIN II detector arrived at LBL just after Christmas 1992, and was installed in our underground Oroville Facility in January 1993. Although the detector background (BKG) is still changing slowly (decreasing), it has already been adequately characterized to enable use of the system for measurements with much greater sensitivity than was possible with the original MERLIN system. General characteristics of the new detector BKG, compared to the original MERLIN system, are as follows:

1) All visible peaks in the U-series and Th-series are at about the same intensity (c/min), except for the Pb-210 peak which is about 8-fold higher than before. The K-40 peak, although still very small, is about 4-fold higher than before.
2) The continuum is about 2-fold higher than before in the 100-300 KeV region, crosses over at about 500 KeV and is lower than before at all higher energies.

Almost all the peaks of interest remaining in the LDEF samples are at energies above 500 KeV. These favorable BKG characteristics, combined with the roughly 4-fold increase in efficiency, translates into at least a 4-fold improvement in our capability to measure the very weak activities in these relatively small-size samples.

MERLIN II Detection Sensitivity

The present shield provides a 7-in square by 18-in high internal volume in which the 4-in diameter by 8-in long vertically oriented detector assembly is centered. There is a 7-in square by 8-in high annular space around the detector barrel. A 3-in high by 7-in square free space also exists above the flat face of the endcap. Literally all shapes and sizes of objects/materials that will fit inside the MERLIN shield are candidate samples. Each different configuration may require separate "calibration" runs with known quantities of appropriate radionuclides in matching matrix materials. Such calibration procedures are often quite time-consuming, but are essential for the frequent special cases that cannot be forced into one of the several "standard sample" formats.

Several representative sample formats of the total available range illustrate the detection sensitivity of the new system; these examples can then serve to guide evaluation of potential applications. The comparisons are based on detection of the 1461 KeV gamma-ray from K-40 in KCl or natural materials, and should be interpreted as mainly "geometric" factors: they do not take into account absorption of gamma-rays inside the sample, as will occur with greater sample thickness or higher-Z sample material, or when measuring peaks from lower-energy gamma-rays. The three examples are:

1) Small (2-in diam.) thin sample centered on the flat face of the endcap (maximum sensitivity);
2) Thin layer (about 1/2" thick) around most of cylindrical endcap, including the flat end;
3) Thick layer (1 1/2" to 2" thick) surrounding cylindrical endcap, with 2-3" thick full-diameter layer at end (maximum sample size).

Approximate values for the detector response to K-40 in KCl, NaCl, or common crustal Earth materials, along with the corresponding minimum detectable potassium concentrations, are as follows:

SAMPLE FORMAT	WEIGHT GRAMS	RESPONSE C/MIN/GRAM	MINIMUM DETECTABLE CONCENTRATION
1. (Small/thin)	20	10.	3 ppm
2. (Thin/extensive)	1000	5.	0.1 ppm
3. (Thick/maximum)	6000	2.5	0.04 ppm

Values in the column labeled RESPONSE represent c/min per gram of natural potassium present in the sample material. Values in the column labelled MINIMUM DETECTABLE CONCENTRATION have the following specific definition: they are concentrations equal to one Standard Deviation on the measured BKG in the K-40 peak, and reflect the statistical quality of data accumulated over a one-week period. Another way to interpret these values is to consider them the highest concentrations of the radionuclide that might just escape detection. Reliable measured values should be obtainable from concentrations two to three times these limiting values.

Similarly defined limits for detection of the two natural radionuclide series (at secular equilibrium), using the largest size sample are:

U-series:	about 20 ppt (parts per trillion)
Th-series:	about 100 ppt.

While longer counting times can provide some improvement (reduction) in the lower detection limit, it would require a one-month count to achieve a 2-fold reduction from the results of a one week counting time. Larger samples (in a larger cavity inside the shield) can also achieve a reduction in this lower limit without extending the counting time - - perhaps a factor of 2 is achievable. However, there does not appear to be a practical route to 10-fold reduction in lower detection limit with our single MERLIN II detector.

The small-sample sensitivity has been determined from a known quantity of KCL chemical in one of our "standard" plastic boxes, which sample was also used with MERLIN I. The container dimensions are a good match to the 2-in square LDEF samples, and it is counted in the same position used for the LDEF samples. The ratio of count-rates in the K-40 (1461 KeV) peak, taken in the sense (MERLIN II)/(MERLIN I), provides a direct measurement of the relative improvement in sensitivity for single-emission (non-cascade) gamma-rays that accompany radionuclide decay. The ratio was measured to be 3.94 for this "maximum sensitivity" position.

Analysis of LDEF NICKEL Samples with MERLIN II

All four NICKEL intentional samples were re-analyzed at LBL in order to measure their Co-60 content with greater accuracy than had been achieved in previous measurements. The samples were counted with the MERLIN II system in June 1993 giving the following results:

SAMPLE	OBSERVED C/MIN	ACTIVITY C/MIN-KG
Nickel #4	0.0551±0.0037	1.05±0.05
Nickel G-12	0.0520±0.0026	1.06±0.03
Nickel I-C3	0.0688±0.0028	1.40±0.06
Nickel Bars(2)	0.0402±0.0022	1.13±0.06

Each sample was counted for the nominal one week period at the highest sensitivity position: centered on the detector endcap. The OBSERVED count rates are derived from the summed contributions of both peaks (1173 Kev plus 1332 Kev). The ACTIVITY values, expressed as c//min-Kg, have been decay-corrected to 1/20/90, the date of LDEF's landing at Edwards Air Force Base.

The above measurements can be linked directly to the previous analyses done in 1990 with MERLIN I: the NICKEL #4 sample was counted in identical "geometries" with both detectors. The Co-60 activity measured with MERLIN I was:

Nickel #4:	Co-60 (2-peak sum)	=	0.373±0.027 c/min-Kg
Ratio:	(MERLIN II/MERLIN I)	=	2.82 ±0.26

The sensitivity increase realized with MERLIN II for Co-60 is smaller than the factor measured with the single gamma-ray that accompanies K-40 decay. The major factor contributing to this difference is related to the emission of the Co-60 gamma-rays in coincidence, which means that as detector size increases, the summing effect becomes greater (the peaks are "robbed" in a greater fraction of all detected events).

Although the primary objective has been to obtain more accurate values for the Co-60 content of these samples, we have also been able to make accurate measurements of the Co-57 and Mn-54 activities that remain nearly 3 1/2 years after retrieval of the satellite. In addition, the presence of Na-22 was detected, although at a level with much lower statistical significance. Searches for Ar-42 (33 year halflife) and Ti-44 (47 year halflife) in each sample were unsuccessful (the possibility remains that these isotopes might show up were all four samples counted at the same time). No other gamma-emitting radionuclides were observed.

LONG-LIVED ACTIVITIES IN TRUNNION SLICES

A group of 20 slices from trunnion sections LHG and RHG were counted together with MERLIN II in an effort to achieve more accurate values for the long-lived radionuclides that were just barely detected in our previous work with the MERLIN I detector.
This composite sample consisted of slices 2 through 6 from sets LHG-N, LHG-S, RHG-N, and RHG-S. Sixteen pieces (in two-layer sets) were arranged around the cylindrical surface of the detector endcap, while the four thickest pieces were placed on the flat face of the endcap. Total sample weight was about 1 Kg.

The composite sample was counted for 17735 minutes during the interval 9/9-23/93. Figure 1 shows several peaks in a portion of the spectrum, including very small (but quantitatively useful) peaks from Ti-44 (47-year halflife) and Na-22 (2.6-year halflife). The two dominant peaks in this energy region belong to Co-60 (5.3-year halflife). Listed below are net count rates observed in the signature peaks that are used to measure the induced-activity isotopes found in the composite sample. No absolute efficiency calibrations have been generated for this sample array, because various members of the array contain different activity levels. However, comparisons of relative activity levels can be made through use of decay-corrections to the values given here. For all but Co-57, self-absorption effects can be ignored for these comparisons.

ISOTOPE	ENERGY KEV	OBSERVED NET C/MIN
Mn-54	834	0.442±0.005
Ti-44	1157	0.0052±0.0007
Co-57	122	0.064±0.003
Co-60	1173	0.063±0.002
	1332	0.062±0.002
	BOTH	0.125±0.003
Na-22	1274	0.0125±0.0009
Ar-42	1524	±0.0004

The entry for Ar-42 is derived from this spectral data and represents a single Standard Deviation on a 5-Kev wide interval of the continuum centered at the energy of the 1524 Kev peak. It may be considered an upper limit for the presence of this isotope.

RADIOACTIVITIES IN LDEF PB BALLAST SLICES

Several Pb-ballast slices cut from parts of the satellite's position stabilization assembly have been analyzed recently with our MERLIN II spectrometer, more than 3 years after recovery of the LDEF. The samples analyzed here are 1/4" thick slices which were "shielded" from space by 3/16" of aluminum and 1/8" of Pb.

The only cosmogenically produced radionuclide we can detect from a week-long count on a single 2" x 2" x 1/4" thick Pb square after so long a decay time is the 33-year halflife isotope Bi-207. This nuclide must have been produced mainly through proton reactions on the stable Pb isotopes Pb-207 and Pb-208. The presence of the shielding mentioned above implies a minimum proton energy of about 60 MeV to produce Bi-207 from a reaction on Pb-207.

Each sample was counted for the nominal one week period at the highest sensitivity position: centered on the detector endcap. The OBSERVED count rates and ESTIMATED activities in pCi/Kg are tabulated below, wherein we have used both prominent gamma-ray peaks to calculate the estimated Bi-207 activities:

SAMPLE	PEAK ENERGY	OBSERVED C/MIN	ESTIMATED pCi/Kg
Pb B-8-916-A-3	570 Kev	0.0442±0.0026	4.9±0.3
	1064 Kev	0.0336±0.0021	6.3±0.4
Pb B-8-916-B-3	570 Kev	0.0506±0.0024	5.6±0.3
	1064 Kev	0.0310±0.0017	5.8±0.3
Pb B-8-920-A-3	570 Kev	0.0675±0.0027	7.5±0.3
	1064 Kev	0.0419±0.0020	7.9±0.4
Pb B-8-920-B-3	570 Kev	0.0663±0.0030	7.4±0.3
	1064 Kev	0.0451±0.0023	8.5±0.4

No decay corrections have been made to these results, in view of the long halflife (33 yr.) of Bi-207 compared to the time since recovery of the LDEF. There may be a relatively small systematic difference between activity values obtained from the two different peaks appearing in the spectrum from each sample, possibly arising from errors in determination of either detection efficiencies or self-absorption in these thick high-Z samples. However, the results have the consistency to support some conclusions:

1). Slices 916-A and 916-B have equal Bi-207 activities;
2). Slices 920-A and 920-B have equal bi-207 activities;
3). The 920- slices have about 1.4 times greater Bi-207 activity than do the 916- slices.

Additional information on the very low-intensity activities was obtained from a composite sample consisting of three 2-in square by 1/4" thick Pb slices and a stacked pair of 2-in square by 1/8" thick Pb slices that were also counted for a week-long period. The two Bi-207 peaks (570 and 1064 Kev) showed a 3-fold increase in intensity compared to results from counting each 1/4" thick slice separately. Several additional low-intensity peaks were also observed from the approximately 720-gram total mass of Pb. These peaks are listed below by energy, along with the isotopes believed to be responsible for their presence:

ENERGY KEV	OBSERVED C/MIN	PARENT ISOTOPE	PARENT HALFLIFE	OBSERVED ISOTOPE
328	0.0091±0.0024	Hg-194	520 y	Au-194
355	0.0060±0.0022	Ba-133	10.5 y	Ba-133
834	0.0023±0.0009	Mn-54	312 d	Mn-54
885	0.0021±0.0008	Ag-110m	270 d	Ag-110m
1094	0.0046±0.0008	Hf-172	1.9 y	Lu-172
1173 1332	(both peaks) 0.0104±0.0013	Co-60	5.3 y	Co-60

Although the observed peak intensities are near the limits of detectablilty, the peaks are believed to be real - - and they do not exist in the BKG, except for Co-60 where the summed BKG peaks have intensity 0.0070 c/min. They represent nuclear reactions requiring that the incident particles (mainly protons) have energies in the hundreds of MeV range. The outcome of this measurement again emphasizes the value of having large mass samples for analysis. Pb samples up to 20 times more massive could easily be accommodated in the MERLIN II system, to produce results considerably more precise than are reported here.

Figure 2 shows a portion of the spectral data, including a small (but quantitatively useful) peak at 1093 KeV which verifies the presence of Hf-172 in the sample, an isotope produced in a reaction requiring a few hundred MeV incident particle energy. The nearby (1120 KeV, Bi-214) peak is a BKG peak, while the adjacent elevated structure to the right is the slowly decreasing evidence for Zn-65 in the detector itself (produced by interactions of cosmic-ray particles with the detector during its existence above ground). The dominant (offscale) peak at energy 1063 KeV is the upper of the two most intense peaks from Bi-207 (the other, at 570 KeV).

USE OF THE MERLIN II SYSTEM FOR SHORT-DURATION MISSIONS

The very high sensitivity and low BKG of our MERLIN II system can also be applied to analysis of shorter-lived radionuclides, such as would be appropriate for measurement from flights much shorter than the LDEF Mission: week-long Space Shuttle missions, for example. The enhanced detection capability, when applied to activities produced in a select set of larger samples, would permit acquisition of the same kind of information from the short-duration flights as has been obtained from long-lived radionuclides produced on the LDEF mission.

A special quick-recovery package could be designed for Space Shuttle missions which, when landed at Edwards Air Force Base, could be at the Oroville Facility and ready for counting within 12 hours of touchdown - - without further transport by air. This procedure would establish feasibility for use one of the most convenient threshold reactions, the production of 15-hour halflife Na-24 in various target elements. When target elements such as Mg, Al, Si, Ca, Ti, Mn, Fe, Co, Ni, and Cu are used, we are availed of reaction thresholds ranging from a few MeV to a few hundred Mev. Many other target/reaction combinations exist, leading to radionuclides having favorable decay schemes with halflives ranging from tens of hours to tens of days, and are eminently suitable for shuttle-length missions.

Implementation of this kind of program on frequent Space Shuttle missions would add significantly to our understanding of both the intensity and short-term variability of the radiation field encountered in low Earth-orbit. Results obtained from such a program would be immediately useful in development (or confirmation) of the planning for missions in which both humans and sensitive instruments are expected to perform for long periods of time - - months to years, in low Earth-orbit.

SUMMARY

We have discussed the high detection efficiencies and low BKG characteristics of the new MERLIN II gamma-ray spectrometry system, installed in January 1993 at the LBL Oroville Low Background Facility. The MERLIN II system is in continuous operation, and is available on a part-time basis to qualified users. We have described analysis of samples from the LDEF satellite, to demonstrate the importance of using such a system to measure the tiny amounts (in the pCi/Kg domain) of induced-activity radionuclides in materials recovered from space missions. Measurement of these radionuclides provides important information on the integrated radiation "exposures" encountered during the missions. Although the LDEF experience relates to long-duration missions, suitable sets of elements can be assembled to provide similar integrals for radiation "exposures" encountered on much shorter duration flights, such as Space Shuttle missions.

ACKNOWLEDGMENTS

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Our appreciation continues for the superb support in detector and electronics technology afforded by the LBL group formerly headed by Fred Goulding, particularly in regards creation and maintenance of our MERLIN spectrometer systems. We also thank R.J. McDonald, newest member of the Low Background Facility team, for his contribution to data analysis for this presentation. And - -thanks again to Kevin Hurley of the U.C. Space Science Laboratory, whose phone call alerted us to this opportunity of a lifetime: participation in the LDEF analysis program.

We respectfully dedicate all our efforts in the LDEF analysis program to the last crew of the Challenger, lost at launch in early 1986, but long remembered in the annals of mankind's journey into space.

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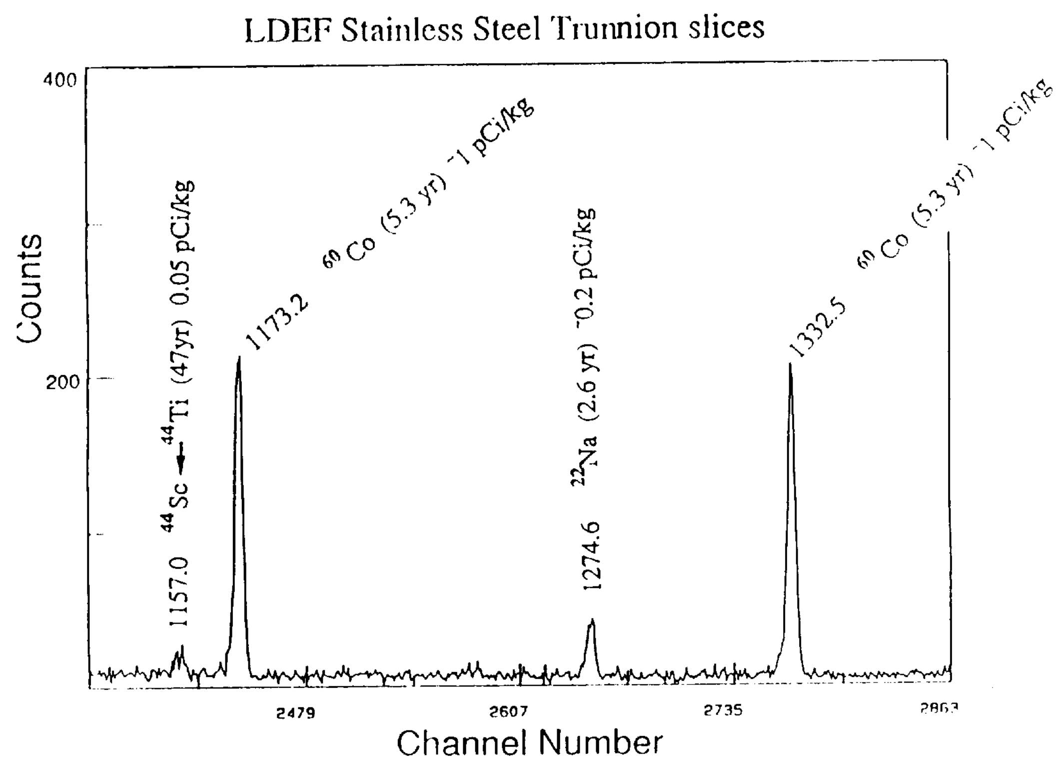

Figure 1. Section of MERLIN II spectrometric data from a 20-pc array of LDEF stainless steel Trunnion Slices, showing peaks in the gamma-ray energy interval 1100-1400 Kev.

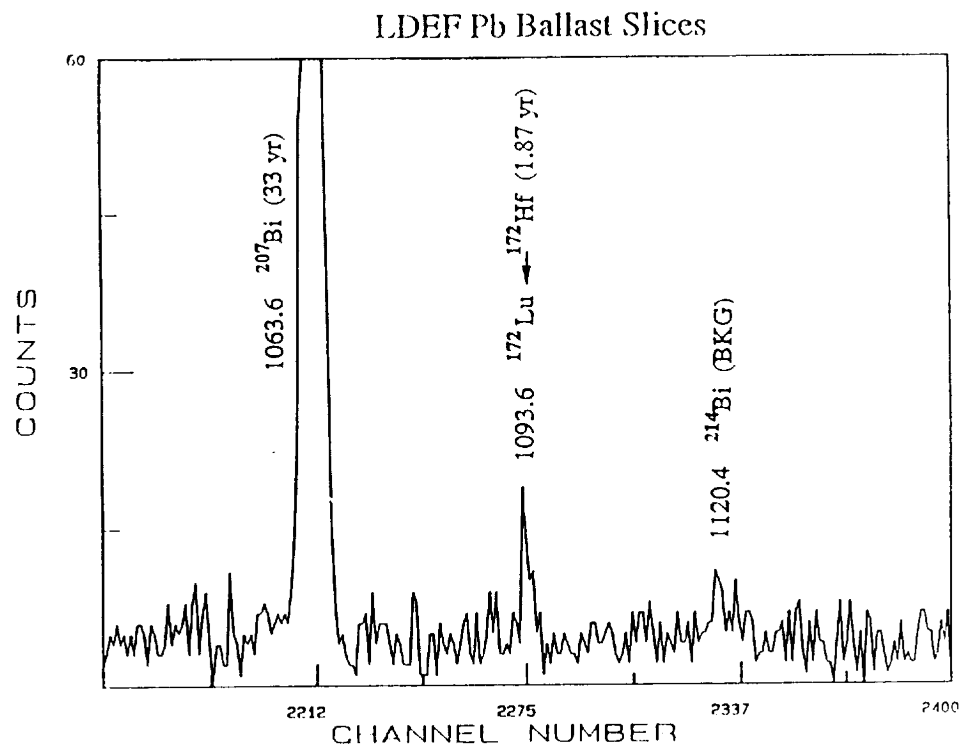

Figure 2. Section of MERLIN II spectrometric data from a 5-pc array of LDEF Pb Ballast Slices, showing peaks in the vicinity of 1100 Kev gamma-ray energy.