LBL-31188

Lawrence Berkeley Laboratory
UNIVERSITY OF CALIFORNIA

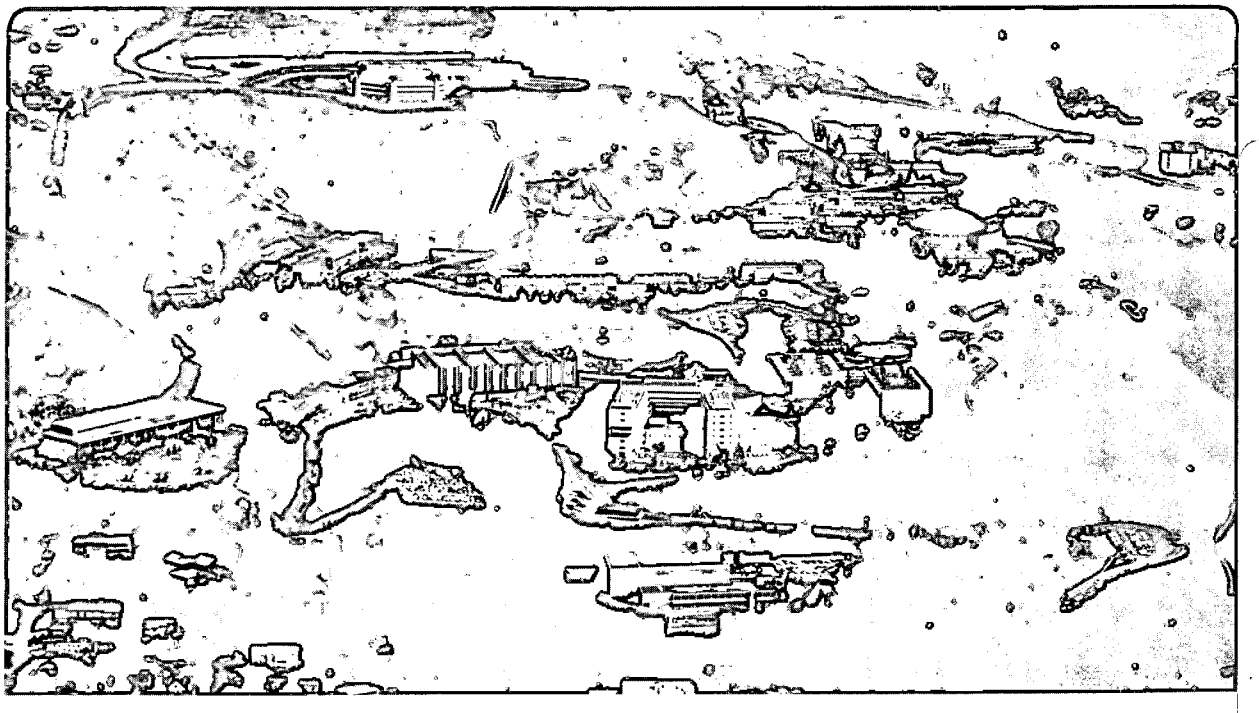
Engineering Division

Presented at the First LDEF Post-Retrieval Symposium, Kissimmee, FL, June 2–8, 1991, and to be published in the Proceedings

Radioactivities of Long Duration Exposure Facility (LDEF) Materials: Baggage and Bonanzas

A.R. Smith and D.L. Hurley

August 1991



LOAN COPY
Circulates
for 4 weeks
Bldg. 50 Library.
LBL-31188
Copy 2

Prepared for the U.S. Department of Energy under Contract Number DE-AC03-76SF00098

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

LBL-31188

RADIOACTIVITIES OF LONG DURATION EXPOSURE FACILITY (LDEF) MATERIALS: BAGGAGE AND BONANZAS

Alan R. Smith and Donna L. Hurley
Lawrence Berkeley Laboratory
University of California
Berkeley, CA 94720

This work was supported by NASA under Marshall Flight Center Order No. H-06815D through U.S. Department of Energy under Contract No.DE-AC03-76SF00098.

RADIOACTIVITIES OF LONG DURATION EXPOSURE FACILITY (LDEF) MATERIALS: BAGGAGE AND BONANZAS

Alan R. Smith and Donna L. Hurley
Engineering Division
Lawrence Berkeley Laboratory
University of California
Berkeley, CA 94720
Phone (415) 486-5679 FAX (415) 486-4122

ABSTRACT

Radioactivities in materials onboard the returned Long Duration Exposure Facility (LDEF) satellite have been studied by a variety of techniques. Among the most powerful is low-background Ge-semiconductor detector gamma-ray spectrometry, illustrated here by results obtained at the Lawrence Berkeley Laboratory's (LBL) Low Background Facilities, in a multi-laboratory collaboration coordinated by Dr. Thomas Parnell's team at the Marshall Spacecraft Center, Huntsville, Alabama.

The observed radioactivities are of two origins: those radionuclides produced by nuclear reactions with the radiation field in orbit; and, radionuclides present initially as "contaminants" in materials used for construction of the spacecraft and experimental assemblies. In the first category are experiment-related monitor foils and tomato seeds, and such spacecraft materials as aluminum, stainless steel, and titanium. In the second category are aluminum, beryllium, titanium, vanadium, and some special glasses.

INTRODUCTION

The voyage of the Long Duration of Exposure Facility (LDEF) in low earth orbit was unexpectedly extended to nearly 6 years -- several years longer than originally planned. This circumstance greatly enhanced the opportunity for post-flight measurement of induced-activity radionuclides created in onboard samples and spacecraft materials. Our earthbound task is to measure these minute quantities of radioactivity -- as many as possible and as accurately as we are able, to insure the description of the radiation field calculated from these measurements will reflect the reality of LDEF experience. Since the quantities of radioactivity to be measured are so small compared to normally encountered terrestial intensities, we must apply the most advanced techniques in radiation detection, in particular: detectors with very high sensitivity and energy selectivity, operated in environments that provide the lowest possible interference (background) with respect to the signals we seek to record.

In the broader context, participation in the LDEF analysis program is truly an opportunity of a lifetime. We consider it a rare privilege to be a part of this worldwide team, and an honor to contribute this report to the Symposium Proceedings.

DETECTOR SYSTEMS AND EXPERIMENTAL PROCEDURES

At LBL, "low-background" radiometric analysis is done at two facilities, one at Berkeley and one at Oroville, using three high-resolution γ-ray spectrometer systems based on high-purity intrinsic germanium semiconductor detectors. All three detectors are of the closed-end coaxial type, with "rated" efficiencies of about 30% -- relative to the efficiency of a 3-in. diameter by 3-in length NaI (Tl) scintillation crystal detector for the 661 KeV γ-rays from Cs-137.

The GEM and NGEM systems are at the Berkeley Low Background Facility (a large room-size facility shielded by 5 feet of low-activity concrete), and use p-type germanium detectors. The GEM detector has a passive Pb shield, while the NGEM detector has a passive Pb shield that is surrounded by an active cosmic-ray veto "shield". Data are collected in the format of 8192-channel multi-channel analyser (MCA) spectra, usually spanning the energy range 35 - 3600 KeV. The MERLIN system has a passive Pb shield, and is located under 600 feet of bedrock at the LBL Oroville Facility. Data are collected in the format of 4096-channel spectra, usually spanning the energy range 15 - 3300 KeV.

The background (BKG) spectra of our Ge-detector γ-ray spectrometers exhibit two distinctly different characteristics: relatively featureless and slowly varying continuous distributions that extend across the entire energy range, and well-isolated peaks superimposed on this continuum. The sharp peaks contain all the information used here for identification and quantification of radionuclides. It is a fortunate circumstance that induced-activity peaks we need to measure rarely overlap peaks in the BKG distributions; hence, our sought-after peaks are usually measured only against the continuum component of the BKG.

Representative values of BKG continuum counting rates are listed below for the three LBL systems. Count rates are given for several energies, in units of counts per minute in a 5-KeV wide interval of suitable width for measurement of a small peak:

	5-KeV Wide Interval		
Energy KeV	GEM c/min	NGEM c/min	MERLIN c/min
500	0.15	0.04	0.015
1000	0.045	0.012	0.004
1500	0.022	0.008	0.002
2000	0.016	0.0035	0.0006
3000	0.0077	0.0014	0.00023

The MERLIN system at Oroville has by far the lowest background of any system available for these measurements, and so is the "star" in our LDEF sample analysis program. It always provides the most accurate results on measurement of the smallest peaks; hence, samples that required measurements for both the lowest intensity peaks and the most comprehensive radionuclide inventories were analysed with the MERLIN system.

Sample analysis times ranged from as short as a few 10's of minutes for some of the system efficiency calibration runs to as long as 10000 minutes for runs on the lowest-activity LDEF samples. Samples were almost always counted at a position as close as possible to the detector -- directly on the flat end face of the detector vacuum vessel, a distance of 6 to 10 mm (detector-dependent).

The tabulation of results appearing in the following sections includes the counting rates observed from the diagnostic peaks used for assay of each radionuclide, and estimates for the decay-corrected absolute activity of each radionuclide, in units of pico-curies per kilogram (pCi/Kg) of metal. The count rates represent net peak areas obtained directly from spectral data in units of counts per minute (c/min), with associated uncertainties of one standard deviation (S.D.) that are based only on the statistics of counting data. Each net peak area was determined through an operator-controlled MCA-resident algorithm: the area equals the difference between the summed counts in the peak-containing interval and a linearly interpolated continuum whose magnitude is determined from the interval endpoints. No use was made of algorithms that employ channel-by-channal peak-shape fitting.

Calculations for absolute radionuclide activities are based on nuclear paramaters given in the most recent edition of Table of Isotopes (Ref 1). Conversion of peak count rates to absolute activity values also incorporate empirically determined parameters for detection efficiency and combined geometric/absorption corrections for the three spectrometer systems used in these measurements. Any summing effects that may occur in detectors from cascade (coincident) γ-rays are NOT taken into account in these preliminary analyses. The calculated activity values have NOT been adjusted to conform with any assumptions relating the degree of secular equilibrium between in-orbit activating particle flux and the activity level existing at satellite recovery time. The only time-domain correction applied was to account for the decay of each radionuclide between recovery time and the time of γ-ray spectrometric analysis.

The evaluation of uranium-series disequilibrium observed in some materials was greatly facilitated through use of the comprehensive lists of γ-rays from this decay chain assembled in the early 1970's by Smith and Wollenberg (ref 2). These lists detail the three decay chains (U-238), U-235, and Th-232), giving energies and intensities appropriate to equilibrium conditions for all known γ-rays in each chain.

RESULTS OF GAMMA-RAY SPECTROMETRIC MEASUREMENTS

Metal Radioactivation Monitors

Among the "Intentional Samples" onboard the LDEF satellite were four sets of five different metal "foils", specially selected for their nuclear reaction properties. Radioactivities (with appropriately long halflives) induced in these materials during spaceflight would be measured after satellite recovery, to provide valuable insight on characteristics of the integrated radiation exposure received in orbit. The selected metals were cobalt, indium, nickel, tantalum, and vanadium (Co, In, Ni, Ta, V), in the form of 2-inch squares of 1/8" thickness. All sets of metals were recovered and later distributed to several of the participating laboratories for radiometric analysis.

We initially received the 4-member set from Experiment P0006 (on the same tray as the tomato seeds); there was no cobalt square in this set. Later in 1990 we received the three other indium squares, so to permit analysis of all 4 members of this element at the same laboratory. We also received the three cobalt members in July 1991, and are presently measuring their radioactive content.

Reported here are quantitative results fore the major γ-emitting radionuclides found in all four members of the Experiment P0006 set, all four indium members, and two of the three cobalt members. Tabulations include the counting rates observed from the diagnostic peaks used for assay of each radionuclide, and estimates for the decay-corrected absolute activity of each radionuclide in units of pico-curies per kilogram (pCi/Kg) of metal. The count rates listed represent net peak areas obtained directly from spectral data in units of counts per minute (c/min), with associated uncertainties of one standard deviation (S.D.) that are based only on the statistics of counting data.

Cobalt Metal Monitors

The three cobalt members were received in July 1991, and are presently being analysed. Preliminary results from two cobalt squares are included here. (Note, there was no cobalt in the Experiment P0006 set.) No evidence was found for the presence of radioactivity brought in this material from earth. Note however, the same suite of Uranium-series radionuclides observed in Vanadium Square #4 and the titanium alloy clips (see elsewhere, this report) were observed in "reactor grade" cobalt samples obtained at LBL in the early 1960's. The following tabulation summarizes results obtained from 10000-minute runs on the GEM and NGEM system at our Berkeley facility.

				Observed		Activity	
Sample	Nuclide	Obs. Peaks	Diagnostic Peaks (KeV)	Net c/min	S.D.	pCi/Kg	S.D.
Co #C9	Mn-54	1	835	0.143	0.004	40.8	1.1
	Co-57	2	122	0.772	0.010	124.9	1.6
	Co-60	2	1173+1332	0.141	0.004	18.6	0.5
Co #G12	Mn-54	1	835	0.076	0.004	28.0	1.4
	Co-57	2	122	0.404	0.008	83.6	1.7
	Co-60	2	1173+1332	0.118	0.004	20.0	0.7

All three radionuclides observed in both cobalt squares were measured with good statistical precision. The activities of Co-60, a product of slow-neutron capture, were seen to be nearly the same in both samples. However, the activities of Mn-54 and Co-57, products of energetic-particle reactions, were about 1.5 times greater in Co #C9 than in Co #G12. This ratio is similar to the north/south ratio for activities measured in near-surface trunnion slices (see the section on Trunnion Slice Activities).

Indium Metal Monitors

The γ-ray spectral data from Indium Square #4 were acquired in late March 1990, about 2 months after LDEF recovery, and contained peaks from seven different space-produced radionuclides (Co-60, Y-88, Rh-101, Rh-102, Ag-110m, Sn-113, and In-114m). Data were also acquired from the indium members belonging to the other three monitor sets, received at a later time and analysed during the period June-July 1990. Space-produced radionuclides observed in these three samples included Rh-102, Ag-110m, Sn-113, and In-114m. No evidence was found for the presence of radioactivity brought in this material from earth.

Each sample was counted for approximately a one-week period. The MERLIN system was used to analyse samples #4 (Experiment P0006), C9 (IC9-IN), and Bars. Indium G12 (G-12-B-3-F) was analysed with the NGEM system at Berkeley. Results are tabulated below for the four radionuclides Rh-102, Ag-110m, Sn-113, and In-441m.

				Observed		Activity	
Sample	Nuclide	Obs. Peaks	Diagnostic Peaks (KeV)	Net c/min	S.D.	pCi/Kg	S.D.
In #4	Rh-102	5	475	0.010	0.004	2.2	0.9
	Ag-110m	5	657+884	0.026	0.005	5.1	1.0
	Sn-113	1	392	0.121	0.008	54.0	3.6
	In-114m	1	191	0.042	0.008	105.	20.
In #C9	Rh-102		475	0.013	0.002	3.2	0.4
	Ag-110m		657+884	0.016	0.002	3.9	0.5
	Sn-113		392	0.059	0.004	40.9	2.7
	In-114m		191	0.008	0.005	55.	35.
In #G12	Rh-102		475	0.014	0.002	2.3	0.3
	Ag-110m		657+884	0.014	0.002	2.3	0.3
	Sn-113		392	0.047	0.003	21.0	1.2
	In-113m		191	0.008	0.003	35.	15.
In Bars	Rh-102		475	0.006	0.002	2.2	0.6
	Ag-110m		657+884	0.008	0.002	3.2	0.8
	Sn-113		392	0.025	0.003	35.1	4.2
	In-114m		191	0.008	0.005	190.	115.

While the statistical precision is adequate for comparing Sn-113 values, activity values for the other three radionuclides have large uncertainties and are marginally suitable for comparison purposes. Quantitative use of values for relatively short-lived In-114m is not recommended, except possibly from Indium #4, which was received and counted much earlier than were the other three samples.

Nickel Metal Monitor #4 (Experiment P0006)

The γ-ray spectral data from this sample were acquired about 2-1//2 months after LDEF recovery. Many peaks from space-produced radionuclides were present; in addition, there was no evidence for the presence of radioactivity brought in this material from earth. The results tabulated below were obtained from a MERLIN system run of 10476 minutes duration at the end of March 1990.

				Observed		Activity	
Sample	Nuclide	Obs. Peaks	Diagnostic Peaks (KeV)	Net c/min	S.D.	pCi/Kg	S.D.
Ni #4	Sc-46	2	889+1121	0.0065	0.0017	1.6	0.4
	Mn-54	1	835	0.0973	0.0033	27.3	0.9
	Co-56	9	847	0.0714	0.0029	33.2	1.3
	Co-57	2	122	2.098	0.0029	322.	2.
	Co-58	1	811	0.0851	0.0033	41.7	1.6
	Co-60	2	1173+1332	0.0288	0.0021	4.7	0.3

Of the six radionuclides reported here, all have activity values with good statistical precision except for Sc-46. All six radionuclides are products of energetic-particle reactions. In addition, a tiny peak appeared in the spectral data at about 1274 KeV energy, indicating the possible presence of Na-22 at a very low level (about 0.5 pCi/Kg); this nuclide was most likely produced by interactions with the galactic cosmic rays.

Tantalum Metal Monitor #4 (Experiment P0006)

The γ-ray spectral data from Tantalum Square #4 were acquired from a MERLIN run of 8562 minutes duration in late March 1990. The data are rich in observable peaks, although a relatively small number of radionuclides is responsible: the five nuclides Lu-172, Hf-172, Lu-173, Hf-175, and Ta-182 generated at least 40 observable peaks. Results are tabulated below for the four radionuclides Lu-172, Lu-173, Hf-175, and Ta-182.

				Observed		Activity	
Sample	Nuclide	Obs. Peaks	Diagnostic Peaks (KeV)	Net c/min	S.D.	pCi/Kg	S.D.
Ta #4	Lu-172	20	1093	0.128	0.004	35.9	1.1
	Lu-173	1	272	0.107	0.006	161.4	8.3
	Hf-175	1	343	0.121	0.006	36.6	1.9
	Ta-182	18	(5 peaks)	0.307	0.008	90.3	2.3

All four radionuclides listed above were measured with good precision. One of the dominant activities, 115-day halflife Ta-182, was produced by slow neutron capture in Ta-181. Its production mechanism is similar to that for production of Co-60 from Co-59; its activity can be compared to the Co-60 activity in Cobalt Square C9 and G12, given in an earlier section of this report. A simple calculation implies the observed Ta-182 activity is significantly greater (perhaps 2-fold greater) than would be expected from a tantalum monitor located with either of the two cobalt monitors. The proximity of Ta #4 to a substantial quantity of hydrogenous moderator (the tomato seeds, for example) could serve to explain such a circumstance.

All other listed radionuclides were produced by energetic-particle reactions, predominantly energetic protons. The other dominant radionuclide, 6.7-day halflife Lu-172, reflects the presence of its parent, 1.37-year halflife Hf-172; thus its activity actually provides a measure of Hf-172 activity.

We found no evidence for the presence of primordial terrestrial radionuclides in this sample of tantalum. This negative finding is consistent with results obtained at LBL on the "natural" radioactivity of this material, in connection with use of similar-size pieces of tantalum in fast neutron flux integrators over the past 25 years.

Vanadium Metal Monitor #4 (Experiment P0006)

The γ-ray spectral data were acquired 3 months after LDEF recovery, and contained few peaks that originated from space-produced radionuclides. The results tabulated below were obtained from a MERLIN run of 9748 minutes duration in mid-April 1990.

				Observed		Activity	
Sample	Nuclide	Obs. Peaks	Diagnostic Peaks (KeV)	Net c/min	S.D.	pCi/Kg	S.D.
V #4	Sc-46	2	889+1121	0.042	0.003	17.4	1.1
	Uranium Concentration:		U-235 (via U-235)		1.5	0.1 ppm	
			U-238 (via Pa-234m)		1.1	0.2 ppm	

The only space-produced radionuclide measurable at counting time was 84-day halflife Sc-46, produced by energetic-particle reactions, predominantly energetic protons. Many other peaks were present at above-BKG intensities, revealing the presence of "baggage" brought from earth as a consequence of the origin and processing of this metal.

The γ-ray evidence is interpreted to show the presence of uranium at a mass concentration of 1.5 ppm (parts per million), or about 500 pCi/Kg. The U-235/U-238 ratio appears to be normal within statistical accuracy of the data. Late members of the U-238 series (Ra-226 and daughters) are virtually absent. The comparable late members of the U-235 series were not observed; however, their expected intensities based on equilibrium with the observed U-235 concentration, would be obscured by the spectrometer system BKG. (See comments elsewhere in this report concerning the titanium alloy radioactivities.) Several peaks useful in Th-series assay (238, 583,

911, 2614 Kev) are slightly above BKG intensities, indicating a mass concentration of about 0.1 ppm (20 pCi/Kg), with a 30% uncertainty (standard deviation). Th-series disequilibrium cannot be accurately determined from data of such poor precision.

This suite of radionuclides in vanadium has been observed previously at LBL, dating from a sample obtained in 1960. We have observed the same radionuclides in titanium alloy Type 6-4, (discussed elsewhere in this report) and in several other samples of this alloy obtained in the last 5 years as candidate material for low-level counting systems.

Tomato Seeds

Representative samples of tomato seeds (from the SEEDS Experiment, described elsewhere in this Proceedings) retrieved from the LDEF satellite were analysed for long-lived gamma-emitters with LBL's lowest-background γ-spectrometer, the MERLIN system, located underground in the power plant of the Oroville Dam (a facility of the California Department of Water Resources). Four 50-gram sealed-in-plastic packets were received, one from each of Layers A, B, C, D of Canister 3 -- wherein Layer A was least shielded and Layer D most heavily shielded from Space Radiation. Short runs (about 1000 minutes) on the A and D packets failed to disclose any differences in their respective content of radionuclides. All four packets were then counted together to achieve maximum sensitivity, in a run of 6767 minutes duration.

The dominant radionuclide observed in all three runs was K-40, the naturally occurring radioisotope of potassium, present as a consequence of the potassium content of the seeds. Both Be-7 and Na-22 were detected at very low concentrations in this 4-packet run, produced from energetic-particle reactions on carbon, nitrogen, oxygen, and sodium--major chemical elements in the seeds. These findings are summarized as follows:

Potassium (1461 Kev):	2.39 ± 0.01 c/min	$\rightarrow$	5400 pCi/Kg
Be-7 (477 KeV):	0.014 ± 0.002 c/min	$\rightarrow$	16 pCi/Kg
Na-22 (1274 KeV):	0.0095 ± 0.0014 c/min	$\rightarrow$	2 pCi/Kg

The induced activity intensities are totally inconsequential, even in comparison to the (benign) activity of essential potassium. The appropriate LDEF researchers were notified immediately of our radiometric analysis results, to facilitate quick distribution of tomato seeds to the millions of eager young grower-participants.

SPACECRAFT STRUCTURAL MATERIALS

Titanium Alloy Clips

Four sets of titanium alloy Type 6-4 (90% Ti, 6% Al, 4% V) two-piece "clips" (#916AE2, #916A12, #920FE1, #920FI1) were sent to LBL for special study, arriving in late June 1990 and analysed immediately thereafter. The only space-produced radionuclides measurable at analysis time were Na-22, Sc-46, and possibly Ti-44. The observed Sc-46 counting rates (889 and 1121 KeV peaks summed) were in the range 0.1 - 0.2 c/min, which translated into an estimated activity

level of 30 - 40 pCi/Kg at satellite recovery time. (These estimates will be refined for inclusion of more precise values in the later comprehensive report.) The Na-22 activity is estimated at about 4 pCi/Kg, and could have been produced mainly by reactions on the 6% abundant aluminum component, rather than by higher-energy reactions on the 90% abundant titanium component. (See the section on Aluminum Keel Plates, this report.)

Detailed analysis of the γ-ray spectral data reveals the presence of a significant concentration of uranium in this material--in the range 12-14 ppm uranium by weight; in fact, uranium series γ-ray peaks are dominant in these spectra. The γ-ray evidence confirms existence of a "normal" U-235/U-238 ratio. Later members of the U-238 chain, Ra-226 and its γ-emitting daughters are virtually absent; however, the later γ-emitting members of the U-235 chain, Th-227 through Pb-211, are present in concentrations appropriate to the observed U-235 abundance. The thorium series is not present in measurable concentration.

We have analysed a number of pure titanium samples at LBL in the past 10 years; none contained measurable uranium (at a detection limit of about 0.003 ppm), and thorium only in the range 0.01 - 0.05 ppm. Several other samples of titanium alloy 6-4 have also shown the presence of the same radionuclides seen in the LDEF parts,but at lower concentrations. The 6% aluminum component could reasonably contribute only 0.05 - 0.1 ppm thorium and 0.02 - 0.05 ppm uranium to the alloy.

This unusual suite of radionuclides is believed to enter the alloy through the 4% vanadium component. (We have previously observed these radionuclides in samples of metallic vanadium.) Implicitly, there must have been a uranium concentration of about 300 ppm in the vanadium fraction of the alloy. Note that carnotite is a major ore of vanadium, and is also a major ore of uranium. The chemistry of the vanadium recovery process could allow uranium and Ac-227, the 21.6-year halflife parent of the observed U-235 daughters, to come through into the finished product, while at the same time rejecting Ra-226 and thorium.

Whatever the route, the presence of these radionuclides at concentrations of the magnitude observed in the LDEF parts is of serious concern to designers of any spacecraft instrument packages that contain nuclear radiation detection systems whose BKG responses are to be minimized.

Trunnion Section Slices

One of our major efforts was to establish depth profiles for induced activities in sections cut from the 3.25-inch diameter stainless steel trunnions. (See elsewhere, this Proceedings, for description of these spacecraft parts.) Our sample suites included four sets of radially-cut slices from both north-facing and south-facing quadrants of trunnion sections LHG and RHG. The outer faces of slices (N1, S1) are the actual trunnion surface, while the inner faces of slices (N7, S7) represent material at a depth of about 1.0 inches (~20 g/cm^2) below this surface. Slices ranged in thickness from 0.034" to 0.251", and weighed between 19.3 and 75.1 grams. Spatial positions of the slices are as follows:

Slice Number	Depth Interval Inches
1	0.000 - 0.034
2	0.049 - 0.114
3	0.128 - 0.232
4	0.247 - 0.383
5	0.398 - 0.564
6	0.578 - 0.774
7	0.789 - 1.040

All three spectrometer systems were used for this series of measurements, spanning the interval from April 1990 through June 1991, employing sample counting times that ranged from 2400 to 10100 minutes. Results are tabulated here for the two radionuclides (Mn-54 and Co-57) that dominated after the unavoidable decay period of 2 months between the time of satellite recovery and sample availability. Much smaller quantities of the radionuclides Na-22, Sc-46, Co-56, Co-58, and Co-60 were also observed in some of the slices. These results will be included in a later report summarizing all the measurements made at LBL on samples from the LDEF satellite mission.

Observed count rates for the Mn-54 834KeV peak ranged from 0.074 c/min to 0.470 c/min; rates for the Co-57 122 KeV peak ranged from 0.021 c/min to 0.177 c/min. The resulting Mn-54 values are of higher precision than are the Co-57 values, mainly because of the greater peak count rates, but also as a consequence of the lower BKG rate at the higher γ-ray energy.

Tabulated below are calculated active values for both Mn-54 and Co-57, in units of pCi/Kg of sample. The "S.D." values are in terms of a single standard deviation on counting data, as propagated through the calculations, and do not include any estimates of uncertainty in detection efficiency or non-uniformity in sample activity.

Trunnion Slice		Mn-54 pCi/Kg	Mn-54 S.D.	Co-57 pCi/Kg	Co-57 S.D.	Trunnion Slice		Mn54 pCi/Kg	Mn54 S.D.	Co-57 pCi/Kg	Co-57 S.D.
LHG	N1	171.3	6.0	38.0	3.6	RHG	N1		(not available)		
	N2	137.3	5.2	33.2	1.6		N2	85.1	2.6	21.7	1.5
	N3	117.5	3.7	25.0	1.6		N3	84.4	3.2	19.7	2.3
	N4	105.1	2.0	21.8	0.9		N4	75.3	1.7	18.9	1.0
	N5	95.0	2.5	22.9	1.6		N5		(not counted)		
	N6	93.1	3.8	21.6	2.5		N6	73.2	1.7	17.2	1.3
	N7	97.0	3.2	18.5	2.2		N7	65.5	3.1	17.0	3.4
	S7	82.6	2.6	17.1	2.2		S7	62.7	2.8	16.4	2.5
	S6	73.4	2.8	17.1	1.6		S6	61.5	2.3	12.4	1.7
	S5	69.3	2.1	13.4	1.3		S5	65.1	2.1	13.6	1.4
	S4	68.4	2.8	14.0	1.7		S4	59.6	2.0	11.6	1.3
	S3	75.8	2.4	15.3	1.1		S3	70.7	2.4	14.2	1.0
	S2	88.2	2.7	15.9	1.0		S2	84.6	2.4	18.0	1.1
	S1	107.3	4.7	20.9	3.0		S1		(not available)		

The Mn-54 activity values in the north-side profile of trunnion section LHG are higher than values in the south-side profile, where the average for the N/S ratio is about 1.56 for the first four outer layers. Activity values for the N7 and S7 slices suggest there may be a broad low peak of Mn-54 activity located near the center of the trunnion section. We can explain the major features of these profiles in a qualitative sense. The inwardly descending profiles are the result of interactions with solar protons, while the broad centrally located "peak" is associated with additional interactions caused by the buildup of secondaries produced by the very energetic galactic cosmic rays. The north-side and south-side Mn-54 activity profiles for the RHG section are much more similar in magnitude than were observed from the LHG section set. The Co-57 activity profiles from both trunnion sections are similar in shape to their Mn-54 counterparts. However, the lower precision of Co-57 data makes it less appropriate to ascribe the same kind of detailed description as is possible for the Mn-54 profiles.

Co-60 was detected in some of the slices, at count rates in the range 0.003 - 0.006 c/min for each of the two peaks. The data are of relatively low precision, making for large uncertainty in the shapes of activity-vs-depth profiles, and hence difficulty in determining whether this activity is space-produced. The implied Co-60 activity values are on the order of 1 pCi/Kg, and fall within the range of Co-60 content of earth-bound stainless steels analysed at LBL, dating from material obtained in the mid 1960's to the present. Analysis of stainless steel "blanks" traceable to the LDEF trunnions could clarify this situation.

Aluminum Keel Plate Radioactivity

Two aluminum (alloy 6061) Keel Plates (KP-4 and KP-9) were analyzed in early 1991, at which time the only measurable space-produced radionuclide was Na-22. (Analysis of two aluminum alloy Scuff Plate Spacers in late March 1990 also revealed the presence of Be-7, as well as the much longer-lived Na-22.) We also obtained values for the "natural" radioactivity content of the keel plate alloy. Tabulated below are results for both space-produced and "natural" radionuclides:

Sample	Nuclide	Diagnostic Peak (KeV)	Observed c/min	S.D.	Activity pCi/Kg	S.D.
KP-4	Na-22	1274	0.238	0.012	140	7
Kp-9	Na-22	1274	0.194	0.007	86	3

Uranium (U-235)	2 ppm
Ra-226 (Bi-214)	0.02 ppm (U-equivalent)
Thorium (TI-208)	0.355 ppm

The measured uranium-series and thorium-series radioactivities lie within the ranges observed for other pure aluminum and aluminum alloy samples analysed at LBL since the early 1960's. (See next section this report.)

Natural Radioactivity in Spacecraft Materials

We have investigated the "natural" radioactivity of a wide variety of materials at the LBL Low Background Facilities in an ongoing effort since 1960, through application of high-sensitivity γ-ray spectrometric techniques. These studies have almost always been conducted in support of specific requirements of various research programs. The resultant body of information relating to "radioactivity in stuff" is neither comprehensive nor complete, and has usually been passed bit by bit to those who requested specific analyses, and without explicit formal publication. Even so, generalities can be gleaned from the 30-year accumulation in this radioactivity "lore" bank that are relevant to the LDEF mission and to the design and construction of future spacecraft.

While the trace radioactivities in engineering materials are usually inconsequential to their intended uses--and hence are not deliberately controlled during production, there are two special cases of concern here in which these radioactivities become very important.

In the first case, their presence in a material interferes with post-flight measurement of radioactivites induced in the material while it was "exposed" to an in-orbit nuclear particle flux, for example: measurement at a terrestrial laboratory of space-produced activities induced in LDEF samples and materials during the satellite's nearly 6-year voyage in low earth orbit. We have already discussed (earlier sections, this report) the presence of uranium-series nuclides in the titanium alloy and pure vanadium, as well as the presence of both uranium-series and thorium-series nuclides in the aluminum alloy. The possibility of earth-borne Co-60 in trunnion stainless steel has been noted.

In the second case, their presence in an instrument and/or its surroundings interferes with measurements the instrument is designed to accomplish in real time during a mission. For γ-ray detection, the radioactivities carried aloft in spacecraft and detector assembly materials (baggage) may contribute significantly to the detector system BKG response, particularly in the matter of identifiable peaks -- the most useful features in γ-ray spectra. It may be necessary to select materials of low intrinsic radioactivity, so to reduce BKG interference with radiation detection mission objectives. The brief discussions of materials that follow are supplementary to comments include earlier, and bear directly on this point.

<u>ALUMINUM</u>: Aluminum and its alloys have been measured to contain uranium at concentrations ranging from 0.3 to 2 ppm; however, Ra-226 and its late-member γ-emitting daughters are virtually absent. Thorium-series γ-emitters are present in the range of 0.05 - 2 ppm equivalent thorium. Note that the γ-ray data does not give direct evidence for the content of Th-232, the parent of the thorium series.

<u>BERYLLIUM</u>: Beryllium has been assayed to contain uranium as the only important radioactive contaminant. Ra-226 and its γ-emitting daughters are virtually absent. Early domestic production (before about 1970) was from beryl ore and yielded metal containing less than 10 ppm uranium. Later domestic production (continuing to the present) from bertrandite ore yielded metal containing 20 - 50 ppm uranium, and recently up to 150 ppm. One sample of metal obtained recently (1989) from China contained about 4 ppm. Also assayed in 1989 was metal from a small domestic "stockpile" of high purity metal, which contained only 1 ppm uranium. Another possible

source of low-uranium metal might be the U.S. National Stockpile, which is at least partly stocked with beryllium recovered from beryl ore.

RARE EARTH OXIDE GLASSES: These glasses are used in high-quality short-focus wide-aperture lens systems, for example: 35-mm single-lens reflex (SLR) cameras of mid-to upper price range. The suite of radionuclides described here has been seen in several SLR lens systems, as well as in samples of "raw" glass. The radionuclides belong to the U-235 series, although U-235 itself is not present. (U-238 is also not present.) The γ-ray evidence confirms presence of 21.6-year halflife Ac-227, the long-lived parent of the observed γ-emitters (Th-227 through Pb-211). Longer-lived Pa-231 may also be present. Uranium concentrations appropriate to the intensities of daughter γ-rays are in the ppm range.

SUMMARY

We have made measurements of in-orbit induced radioactivities and "natural" radioactivities in a number of samples and materials from the LDEF satellite. Success in this effort required the use of state-of-the-art low-background germanium semiconductor detector γ-ray spectrometric technology. Results of our measurements, in combination with results obtained at several other laboratories, will lead to a clearer understanding of the radiation field in which LDEF travelled for nearly 6 years. This information will be useful in design of future spacecraft, such as the space station Freedom. Our LDEF analysis experience can also serve to guide improvements in future efforts of this kind: arranging more suitable earth-bound analysis facilities, as well as selecting the types and quantities of materials sent on spacecraft voyages.

ACKNOWLEDGMENTS

We express our deepest appreciation to all NASA personnel associated with the LDEF mission: in particular, the 1984 Challenger crew who launched the satellite and the 1990 Columbia crew who brought it back to earth; the LDEF mission primary team at Langley, headed by Dr. William Kinard; the Langley team headed by Ms. Arlene Levine, who organized this Symposium on very short notice; and the Special Investigation Group for radioactivity measurements at Huntsville, headed by Dr. Thomas Parnell and including Dr. Gerald Fishman and B. Alan Harmon. Special thanks are due Alan Harmon for his masterful effort at overseeing distribution of samples among participating laboratories on a timely schedule.

We are also appreciative for the superb support afforded by Fred. Goulding's group at LBL in detector and electronics technology, particularly in regards to the creation of our MERLIN spectrometer system. And -- thanks 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 this program to the last crew of the Challenger, lost at launch in early 1986, but long remembered in the annals of man's journey into space.

BIBLIOGRAPHY

1. Lederer, C.M., et al: Table of Isotopes, Seventh Edition, John Wiley and Sons, New York, NY, 1978.

2. Smith, A.R., and Wollenberg, H.A.: High-Resolution Gamma Ray Spectrometry for Laboratory Analysis of the Uranium and Thorium Decay Series, in The Natural Radiation Environment II, CONF-720805-P1, 1972, pp. 181-231.

This work was supported by NASA under Marshall Flight Center Order No. H-06815D through U.S. Department of Energy under Contract No.DE-AC03-76SF00098.

LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
INFORMATION RESOURCES DEPARTMENT
BERKELEY, CALIFORNIA 94720