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RADIOACTIVITIES INDUCED IN SOME LDEF SAMPLES*

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SUMMARY

Radioactivities induced in several LDEF samples were measured by low-level counting at Los Alamos and elsewhere. These radionuclides have activities similar to those observed in meteorites and lunar samples. Some trends were observed in these measurements in terms of profiles in trunnion layers and as a function of radionuclide half-life. Several existing computer codes were used to model the production by the protons trapped in the Earth's radiation belts and by the galactic cosmic rays of some of these radionuclides, ^{54}Mn and ^{57}Co in steel, ^{46}Sc in titanium, and ^{22}Na in alloys of titanium and aluminum. Production rates were also calculated for radionuclides possibly implanted in LDEF, ^{7}Be, ^{10}Be, and ^{14}C. Enhanced concentrations of induced isotopes in the surfaces of trunnion sections relative to their concentrations in the center are caused by the lower-energy protons in the trapped radiation. Secondary neutrons made by high-energy trapped protons and by galactic cosmic rays produce much of the observed radioactivities, especially deep in an object. Comparisons of the observed to calculated activities of several radionuclides with different half-lives indicate that the flux of trapped protons at LDEF decreased significantly at the end of the mission.

INTRODUCTION

Many structural pieces from the Long Duration Exposure Facility (LDEF) were measured using low-level gamma-ray spectroscopy by several research teams (*e.g.*, refs. 1,2.) Several radionuclides were observed, as was predicted on the basis of calculations done using the energetic-particle environment expected at LDEF (ref. 3). Two sources of energetic particles are expected to dominate the production of these induced radioactivities, energetic ($E \sim 10^2$-10^3 MeV) protons trapped in the Earth's radiation belt and the high-energy ($\gtrsim 4$ GeV/nucleon) galactic-cosmic-ray particles that penetrate the Earth's magnetic field and reach LDEF in its 28.5° inclination orbit (ref. 3). There is very little experience with radionuclides made in material in a low-Earth orbit like LDEF's, which is what makes the induced radioactivities measured in LDE

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Measurement of the same isotope in a sequence of successively heavier target elements provides information about increasingly higher-energy components of the radiation field. For example, the reactions to produce Na-22 from Al, Si, S, Ti, and Fe have increasing energy thresholds ranging from a few tens of MeV up to a few hundreds of Mev. Reactions to produce 47-year halflife Ti-44 from Ti, Mn, Zn, and Ge encompasses a similar range of threshold energies. Suites of target elements that produce appropriately "short" halflife radionuclides can also be found to provide a similar range of energy spectral information from short missions. In either case, the enhanced system performance and increased sample size discussed earlier will be required if we are to extract the best quality radiometric information from returned samples.

... LDEF samples, but are attractive

samples so unique and valuable. However, there is much experience with cosmic-ray-produced (cosmogenic) nuclides made by energetic particles in lunar samples (ref. 4), meteorites (ref. 5), and terrestrial samples (ref. 6). We compare our LDEF radioactivities with production rates of cosmogenic nuclides calculated with models developed for these other objects.

MEASURED RADIOACTIVITIES IN LDEF SAMPLES

At Los Alamos, we measured activity-versus-depth profiles in four sets of trunnion layers, six trunnion sections, five parts of aluminum alloy from end support retainer plates, two aluminum alloy keel plate pieces, and two titanium clips, and details on these measurements are in ref. 7. All activities are corrected to 20 January 1990. In the aluminum samples, we observed 2.6-year ^{22}Na with activities ranging from 3.8±0.6 becquerel (disintegrations per second) per kilogram of sample (Bq/kg) to 5.2±0.6 Bq/kg, in good agreement with the ^{22}Na activities of ref. 1 in aluminum tray clamps and of ref. 8 for other keel-plate samples.

As shown in ref. 1, our measured activities for 312-day ^{54}Mn in layers of the D trunnion section, 3-5 Bq/kg, are in good agreement with measurements in adjacent samples done by D. C. Camp at the Lawrence Livermore National Laboratory. A plot of our ^{54}Mn results for layers from the D section of the west trunnion is shown in figure 1, and shows the trend seen in all sets of trunnion layers of a decrease in radioactivity from the surface to the center of the trunnion.

In two titanium clip samples, which we received more than 200 days after LDEF was recovered, we observed low activities of ^{22}Na of 0.6±0.3 and 0.7±0.3 Bq/kg, many gamma rays from uranium and its daughter isotopes, but no clear signal for 84-day ^{46}Sc. Our 3-standard-deviation limits for ^{46}Sc were 3.3 and 4.1 Bq/kg. Our limits for ^{46}Sc are consistent with the activities of 1.1-1.5 Bq/kg estimated for other titanium clips in ref. 8. As ^{46}Sc is made from ^{48}Ti by the same reactions that make ^{54}Mn from ^{56}Fe, it is a little surprising that the measured radioactivity of ^{46}Sc is much lower (by a factor of ~3) than that of ^{54}Mn in the trunnion samples.

MODELS FOR PRODUCTION OF COSMOGENIC NUCLIDES

These radionuclides measured in LDEF pieces have activities that are at roughly the same levels as observed in lunar samples (ref. 4) and meteorites (ref. 5), which range from a few tenths of a becquerel per kilogram for high-energy products like ^{10}Be to ~3 Bq/kg for solar-proton-produced radionuclides like ^{26}Al and ^{22}Na in the very surfaces of lunar rocks. However, most samples from LDEF, like the titanium clips, are very different in composition from extraterrestrial objects, so direct comparisons are difficult to make. Several models have been developed and are well tested for cosmogenic nuclides in lunar samples and meteorites, and we apply two of these models, both from ref. 4, to our LDEF samples. One calculates production rates of nuclides by galactic-cosmic-ray particles. The other was designed for nuclides made by solar-energetic-

slow-neutron capture radionuclides, it is crucial to check for the pre-launch content of these radionuclides, for example: Co-60 in cobalt "intentional" samples. This is especially important when long halflife nuclides are to be measured from short-duration missions, as is the case for Co-60 in cobalt

With improved detection efficiency, increased sample size, and a favorable sample recovery schedule, it is feasible to undertake "frequent flyer" missions on the Space Shuttle with 5- to 10-member sets of "intentional" samples. It is important to build a multi-mission database for this kind of information, initially to test its validity, but mainly to supply important information on characterization of particles, which are often called solar-cosmic-ray (SCR) particles. We also used another computer code system to test the assumptions of the SCR model.

Radionuclide Production by Galactic-Cosmic-Ray Particles

Production of cosmogenic nuclides by the high-energy particles in the GCR have been well reproduced in lunar samples by the GCR model of Reedy and Arnold (ref. 4). This model has also been successfully extended to meteorites (cf., ref. 5). However, this model is not directly applicable to LDEF as the Earth and its magnetic field affect the fluxes of GCR particles reaching LDEF. Only GCR particles arriving from 2π steradians of space reach a point near the Earth, thus production rates calculated for meteorites need to be reduced by a factor of two to be applicable to LDEF, although calculated rates with the lunar model do not need this factor. The Earth's very strong magnetic field scatters away a large fraction of GCR particles for geomagnetic latitudes less than $\approx 60°$. This effect has been known for terrestrial cosmogenic nuclides, such as ^{14}C, and needs to be applied to nuclides made in-situ in surface samples. Production rates near the Earth's equator are about a factor of three lower than those near the geomagnetic poles, while those around a geomagnetic latitude of $\approx 30°$ are lower by a factor of about two (ref. 6). As LDEF's orbit had an inclination of 28.5°, this factor averaged over its orbit is estimated to be about 2.5.

The GCR particle-flux model of ref. 4 was used with the cross sections for the important reactions. Several fluxes for large meteorites and the Moon were used as LDEF pieces were usually on the main part of the spacecraft or, like the trunnions, were fairly thick. GCR production rates for the very surface or in a very small object would be lower as the leakage of particles or reduced production of secondary neutrons lower GCR production rates (refs. 4,5). As described in ref. 4, the cross sections used in this model are for neutrons at energies below a few hundred MeV and mainly for protons at higher energies. Most cross sections used here are similar to those in ref. 4.

The calculated production rates by GCR particles corrected to LDEF's orbit are given in the last column of table I for several materials in LDEF. Other radionuclides observed or being searched for in LDEF pieces include ^{7}Be, ^{10}Be, and ^{14}C; the latter two could be like ^{7}Be and implanted on the surfaces on LDEF's leading side (J. C. Gregory, G. F. Herzog, and A. J. T. Jull, priv. comm.). Production rates of these implanted radionuclides are also given in table I to help show that observed concentrations are more than those expected by GCR-induced nuclear reactions.

For the LDEF mission length of 69 months, all of these radionuclides except ^{22}Na, ^{10}Be, and ^{14}C should have their activities in equilibrium with their production rates. Thus the GCR contribution to the activity of ^{54}Mn in steel should be about 0.9 Bq/kg, which is much lower than the measured activities. The activity for ^{22}Na would be about 90% of its production rate, so its activity in aluminum pieces from the GCR is about 0.5 Bq/kg, again less than observed. The calculated production rate for the "implanted" isotope ^{7}Be is much lower than for those induced

SUMMARY

The perspective for this discussion of the LDEF sample analysis effort is forward-looking: evaluation of our experience at obtaining the reported results in the context of improvements that can be implemented during future missions. Examples have been given to emphasize the need for larger samples, as well as more efficient (larger) detectors. Choices for target elements (intentional samples) have been discussed, as well as the schedules for sample retrieval after spacecraft touchdown. Finally, some applications are outlined for use of induced-activity analysis of materials returned to earth after various times in space, as one of the promising avenues toward identifying (any) temporal variations in

"in-situ," like ^{54}Mn and ^{22}Na, consistent with the interpretation (ref. 9) that the ^{7}Be found on LDEF's leading surface was not produced in LDEF. Production rates for ^{10}Be and ^{14}C are lower than for ^{7}Be, and the calculated GCR production rates for these isotopes in iron are down by a factor of ~2 for ^{10}Be to ~10 for ^{14}C relative to their rates in aluminum.

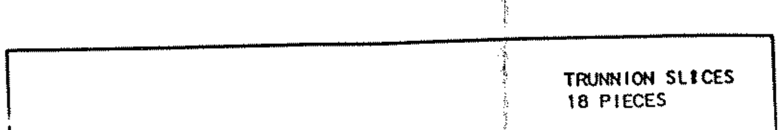
Radionuclide Production by Trapped Protons

The energy spectra of the trapped protons near LDEF's orbit are estimated (ref. 10) to be fairly steeply dropping with energy with most protons having energies of 10^2 to 10^3 MeV. These spectra are similar to those of the energetic SCR protons (ref. 4), although there are relatively more higher-energy protons in the trapped radiation than in the SCR.

Protons with an energy of 100 MeV only have ranges in aluminum and most materials of ≈10 g/cm^2, which is a small fraction of their interaction lengths. The few particles that do interact before they are stopped make few secondary particles because of their relatively low energies. The primary particles themselves induce most reactions. Thus the most important interaction mechanism for such particles is slowing down (and stopping), which occur due to ionization losses along the particles' paths and is a well known process. A model is presented in ref. 4 that involves production of nuclides by particles with energies below a few hundred MeV by considering only ionization energy losses. This model has been successfully applied to many studies of the interactions of SCR protons and alpha particles in the Moon and meteorites (ref. 5).

The trapped protons at LDEF's orbit tend to have relatively more energy than those in the SCR, thus some of the assumptions made above are weakened. To test whether secondary particles made by trapped protons could be important, we used the pair of Monte Carlo codes in the LAHET Code System (LCS) to calculate the interaction of trapped protons with LDEF's trunnion. These codes are LAHET (ref. 11), the Los Alamos High Energy Transport code, and MCNP (ref. 12), the Monte Carlo Neutron Photon code. A solid sphere of radius 4.125 cm with density of 7.62 g/cm^3, the same as the LDEF trunnion, and with the composition of the trunnion 17-4PH steel was used in these codes with the trapped proton flux of ref. 10 for protons traveling in the direction of geographic east at an altitude of 500 km. This proton spectrum is harder than that for geographic west at 500 km or those at 300 km altitude.

The proton and neutron fluxes crossing a surface at a depth of 0.59 cm (4.5 g/cm^2) in this sphere is shown in figure 2. While there is a fairly strong flux of neutrons at lower energies, the flux of protons exceeds the flux of neutrons for energies above about 20 MeV. While the energy where the proton and neutron fluxes cross varies slightly with depth, being at lower energies at the surface and at somewhat higher energies in the center, the trend is similar throughout the trunnion. Using a cylinder instead of a sphere for the trunnion would tend to increase slightly this cross-over energy. These fluxes calculated with LCS show that for the trapped radiation, while secondary neutrons are not negligible, protons are the dominant particle inside a trunnion piece. A very similar conclusion was obtained by the calculations of ref. 3, which showed that production by the protons in the trapped radiation dominates production by neutrons until



depths of ≈25 g/cm^2.

The rates for the production of nuclides by trapped protons in LDEF samples were calculated with proton fluxes calculated inside an object with the SCR model in ref. 4 and cross sections for proton-induced reactions. As with the LCS calculations, the geographic-east flux at 500 km of ref. 10 was used. The ionization parameters used for slowing-down and stopping calculations are those for LDEF's trunnion steel. The SCR code in ref. 4 only considers slab and spherical geometries. Calculations were done using omnidirectional fluxes on solid spheres of radius 31.5 and 63 g/cm^2 and for a semi-infinite slab. The proton fluxes calculated with the slowing-down model of ref. 4 agree well with those from the LCS calculations. At the surface and at a depth of 15 g/cm^2 from the surface, the calculated production rates in the 31.5-g/cm^2 sphere and a slab differed by factors of ≈25% and 2.5, respectively, with the rates at these depths in the 63-g/cm^2-radius sphere intermediate. As a long, narrow piece, like a trunnion, is intermediate between a sphere and a slab, the results for a 63-g/cm^2 sphere are used for the trunnion and for the other geometries.

Most cross sections used for these calculations are similar to those in ref. 4. For two induced radionuclides of interest, ^{54}Mn in steel and ^{46}Sc in titanium, more recent measured cross sections for proton reactions with iron and titanium from refs. 13, 14, and 15 were used. Only cross sections for proton-induced reactions are used.

The flux at 500 km is too high, as LDEF never was that high (having started at 476 km and recovered at 332 km, ref. 16), and fluxes at lower orbits are lower (ref. 10). The calculated production rates for the geographic-west flux at 500 km and for the geographic-east flux at 300 km are about 3–5 and about 50–60 times lower, respectively, than for the geographic-east flux at 500 km. Thus LDEF probably was exposed to a flux of trapped protons that was a factor of several to an order of magnitude or more lower than used here. However, these calculated rates can be used to show that the measured radioactivities are consistent with production by trapped protons.

DISCUSSION

The radioactivities measured in LDEF are similar to those observed in meteorites and lunar samples. Production-rate models developed for extraterrestrial materials have been used to calculate the production rates of several radionuclides in samples of LDEF for particles in the galactic cosmic rays and for protons in the trapped radiation. These calculated production rates are consistent with the measured radioactivities.

Production rates were calculated for four radionuclides measured in three materials from LDEF and for three radionuclides that could have been implanted in LDEF's leading surface. The rates for these implanted isotopes, ^{7}Be, ^{10}Be, and ^{14}C, can be used to help show that they were not made "in-situ" by the GCR or trapped protons. As these three radionuclides are made mainly by high-energy particles, their depth-versus-activity profiles are fairly flat, as the calcu-

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lated rates for trapped protons given in table I show. Thus a sample away from the surface of LDEF can be used to establish the production rate by "in-situ" nuclear reactions for an "implanted" species in LDEF.

The production rates estimated for the GCR are lower than the measured activities by factors of several, although there are a number of uncertainties in going from GCR interactions in meteorites or lunar samples to GCR-induced reactions in LDEF. The production rates by GCR particles vary slowly with depth (ref. 4), and production by GCR particles at depth in LDEF probably is important. The calculated GCR rates are less than the radioactivities measured in LDEF.

As noted above, the production rates calculated here for trapped protons are too high. As these calculated rates are also higher than the observed activities, then the lower fluxes of trapped protons that LDEF was exposed to would be adequate to make most of the induced radioactivity induced in LDEF. The measured radioactivities of ^{54}Mn in the trunnion layers dropped less than a factor of two in going from the surface to a depth of 15 g/cm^2 (a radius of 0.9 inches in figure 1), while the calculated production rates dropped a factor of three over this depth. Thus trapped protons by themselves probably can not account for all of the activity induced in the trunnion. However, the shape of the activity-versus-depth profile clearly shows a surface excess of radioactivity that is similar to that made by relatively-low energy particles.

The calculated production ratios for ^{54}Mn/^{57}Co made in steel by either trapped protons or the GCR are about 6 – 7, similar to their measured activity ratios. The calculated production ratios for ^{54}Mn in steel to ^{22}Na in the aluminum 6061-T6 alloy is about unity, again consistent with the measurements. The fact that these production ratios are similar for both trapped protons and the GCR means that the observed activity ratios can not be used to unfold relative contributions by these two types of incident radiations.

The calculated production ratios for ^{54}Mn in steel to ^{46}Sc in the titanium Ti-6Al-4V alloy is about unity, while the measured activity ratio is about 3 to 1. While shielding of the titanium by overlying material could affect this ratio, there does not appear to have been enough differences due to the locations and shielding of these samples to give a ratio of 3. Another explanation for the lower activity of ^{46}Sc is that it was made nearer to the end of LDEF's mission than was ^{54}Mn. LDEF's orbit was lower then (ref. 10), and the fluxes of trapped protons decrease rapidly in very low orbits (ref. 10).

The calculations present here, while not very detailed, show that the radioactivities induced in LDEF are consistent with our experience in meteorites and lunar samples. There is a component due to the relatively-low-energy protons in the trapped radiation that contributes the observed enhancement in activity at the surface of the trunnions. Another component involves higher-energy particles and their secondary neutrons that contributes a fairly flat profile and much of the production at depth. Calculated production ratios for all isotopes but ^{46}Sc are consistent with measured ratios. Activities of 84-day ^{46}Sc are probably low because of the lower fluxes of trapped protons near the end of LDEF's mission when LDEF was in a very low orbit.

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CHARGED PARTICLE ACTIVATION STUDIES ON THE SURFACE OF LDEF SPACECRAFT

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RESULTS

Seven sample materials, Aluminum, Silicon, Nickel, Copper, Zirconium, Tantalum, and Tungsten, from the LDEF were analyzed for gamma emission by high resolution, high precision gamma ray spectroscopy techniques. The gamma ray spectroscopy equipment is based upon a Nuclear Data Corp. Genie 9900 system. Data acquisition, display, and processing are controlled by a DEC MicroVAX II computer. A Ge(Li) solid state detector was utilized to detect emitted gamma rays. The detector had an efficiency of approximately 20% with a resolution of approximately 1.9 keV (FWHM Co-1332 keV). The overall system operated as a 8192 multichannel analyzer. The gamma ray spectrum ranged from about 100 keV up to about 2 MeV.

Table I. Calculated Production Rates in LDEF Samples.

The target is the material or alloy being considered, such as the titanium alloy used for clips. Rates are calculated for the indicated radionuclide in that target material. The first three rates are for production by trapped protons at different depths, and the last one is for production by GCR particles away from a surface. The incident omnidirectional flux for the trapped protons was the geographic-east 500-km one in ref. 10, and the target was modeled as a sphere of radius 63 g/cm^2. See text for details on the two sets of calculations. The calculated production rates are in units of atoms s^{-1} kg^{-1} for the composition of the indicated target.

Target	Nuclide	Surface	4.5 g/cm^2	15 g/cm^2	GCR
Steel	^{54}Mn	22.5	12.5	7.1	0.9
Steel	^{57}Co	4.5	2.1	1.2	0.13
Ti alloy	^{46}Sc	23.5	13.9	8.2	0.9
Ti alloy	^{22}Na	1.7	1.0	0.6	0.08
Al alloy	^{22}Na	24.7	14.5	8.5	0.7
Aluminum	^{7}Be	1.4	1.0	0.6	0.1
Aluminum	^{10}Be	0.2	0.15	0.1	0.03
Aluminum	^{14}C	0.2	0.15	0.1	0.07

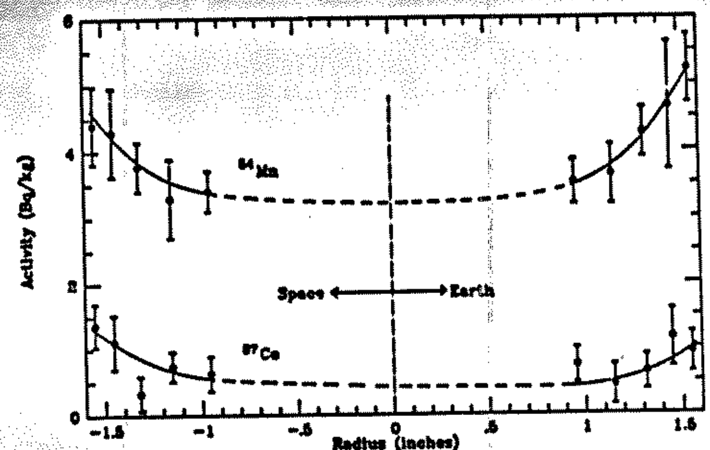

Figure 1. Indu~ed radioactivity in section D of the left hand (west) trunnion. The curves are to guide the eye, especially to the enhanced activity near the surface and the flattening of the profile near the center.

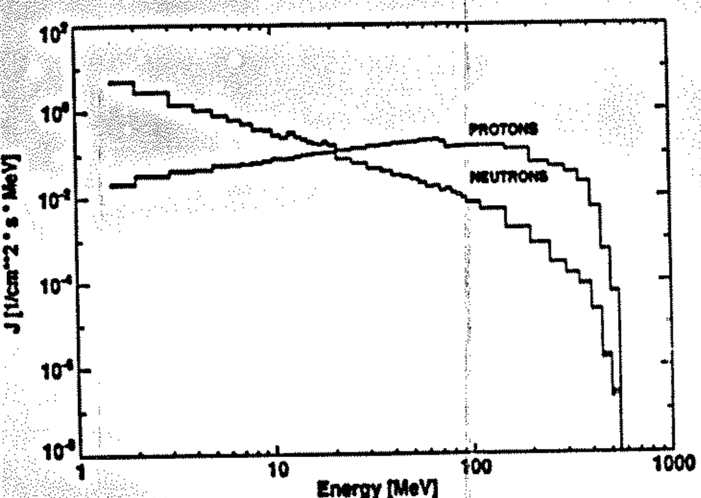

Figure 2. Proton and neutron fluxes calculated by the LAHET Code System as crossing a surface 0.59 cm (4.5 g/cm^2) deep in a steel sphere of radius 4.125 cm (31.5 g/cm^2). The incident flux was the geographic-east, 500-km trapped proton one from ref. 10. The fluxes of protons dominate except for energies below 20 MeV.

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