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STUDIES OF NEUTRON AND PROTON NUCLEAR ACTIVATION IN LOW-EARTH ORBIT

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ABSTRACT

The expected induced radioactivity of experimental material in low-Earth orbit has been studied in regards to the characteristics of activating particles such as cosmic rays, high energy Earth-albedo neutrons, trapped protons, and secondary protons and neutrons. The activation cross-sections for the production of long-lived (half life > 1 day) radioisotopes and other existing nuclear data appropriate to the study of these reactions have been complied. Computer codes required to calculate the expected activation of orbited materials have been studied or developed. A computer code developed in this study has been used to predict the activation by trapped protons of materials placed in the expected orbits of LDEF and Spacelab II. Techniques for unfolding the fluxes of activating particles from the measured activation of orbited materials have been studies.

List of Tables and Figures

*Available upon request from Author.

INTRODUCTION

Many experiments and much experimental equipment placed in near-Earth orbit are sensitive to induced radioactivity. The radiation sources producing this activation are cosmic rays, trapped protons, high-energy Earth-albedo neutrons, and secondary protons and neutrons. When the orbit of a spacecraft passes through regions of trapped protons, these protons and their secondaries will dominate the activation. While the flux of the trapped protons is known to within a factor of two or less [1], the fluxes and the effects of the secondaries are not well understood. The viability of many orbital experiments requires a better understanding of the dominate factors involved in induced radioactivity and the ability to predict the effects of induced radioactivity on these experiments.

In order to better understand the fluxes of these activating particles and the effects of their interaction with spacecraft, NASA has scheduled several experiments in low-Earth orbit. One of these is being planned and carried out by the Astrophysics Branch of SSL at MSFC. Several activation samples are scheduled to be flown on the Long Duration Exposure Facility (LDEF) and on Spacelab II. By measuring the activation of these samples after return from orbit information concerning the fluxes of activating particles is to be obtained.

The unfolding of the particle flux from the measured sample activation requires a thorough knowledge of the cross-sections for the particular reaction under study as well as competing reactions contributing to a particular activation. Also, techniques for unfolding the fluxes from the activation must be known. This report contains the results of a study of the various reactions which can activate the samples, of the cross-sections for these reactions, of the anticipated activation of these samples for the proton flux expected in the planned orbit, and of computer codes needed to calculate the expected activation or to unfold the fluxes of activating particles. Particular emphasis, and calculations, is given to activation produced by primary proton fluxes expected in the LDEF orbit. Suggestions are made for further studies, calculations, and experiments.

LDEF and Spacelab II

It is currently planned to fly five activation samples aboard LDEF and Spacelab II. Due to the shortness of the Spacelab II mission (about one week) little activation of the

Spacelab II samples is expected. However, LDEF is scheduled to be in orbit for a minimum of six months so the activation of the samples aboard it will be more significant.

The LDEF is a structure designed to carry 84 trays into which a large number of passive experiments can be placed. It is a cylindrically shaped structure whose cross-section is a regular twelve-sided polygon with its axes perpendicular to the base. It is to be flown with one base directed towards the Earth at an altitude of about 500km with an orbital inclination of 28.5°.

The five activation samples to be flown on LDEF and Spacelab II consist of 2 inch by 2 inch by 1/8 inch thick slabs of naturally occurring Vanadium, Nickel, Cobalt, Indium and Tantalum. These 99.99 percent elementally pure samples are to be mounted on LDEF experiments A0114, M0001, and M0002.[2] On A0114 the samples are to be mounted inside 3/8 inches of aluminum and/or quartz on a tray to be mounted on the leading and trailing sides of the facility. On M0001 the samples should be cut into 1 x 2 x 1/8 inch segments to be mounted between the four heavy-ion detectors sited on the aphelion side of the spacecraft. On M0002 the samples will be mounted behind a thin honey-combed aluminum plate in a quarter tray sited on the Earth side of the spacecraft. On M0001 and M0002 little degradation of the trapped proton flux is expected because of the covering material but on A0114 considerable degradation is expected. This degradation will not be addressed in this report.

Methods for Predicting the Activation from Trapped Protons

The activation of the samples is caused, first, by the nuclear capture of the primary particles and, second, by the capture of the reactant products. Given sufficient energy, these products would include secondary protons, neutrons, photons, and product nuclei. The first three types of secondaries and some of the product nuclei (deuterons - or pn-, tritons - or p 2n -, He-3-or 2pn -, etc) occur predominantly at low energies; i.e., below about 100 MeV. Compound nuclear reactions are the dominant modes of reaction below 50 MeV followed by direct reactions above 50 MeV with spallation processes increasing in strength above about 100 MeV.

The calculation of the product of a given reaction for a very thin target can be given by

$$P(E) = \frac{N_A}{m} \sigma(E) \phi(E) \rho t A \qquad (1)$$

where N_A is Avogadro's number, m the isotopic mass of the target, p the density of the target, t the target thickness, A the areas of the target, $\sigma(E)$ the reaction cross-section at energy E, and $\phi(E)$ the incident flux (particles per unit time per unit area). However, for thick targets, the incident flux at energy E may be reduced by absorption and energy degradation. The energy degradation of charged particles can be handled with the known stopping powers of the target material. The nuclear absorption comes from the many inelastic reaction channels available at a specific particle energy. Two techniques commonly used to perform such reaction calculations are the nucleon transport codes such as HETC and a straight-forward approach using a modification of Eq. (1) with known cross-sections.

The high energy transport codes HETC [3] which includes evaporation processes in its latest version has been used for incident particles from 15 MeV to 1. TeV. The code assumes an overall geometrical type cross-section and uses Monte Carlo techniques to follow the individual nucleon-nucleon reactions (inside a nucleus) until all primary and secondary reactions cease. The resultant event chain can be interpreted in many ways including the number of radioactive product nuclei produced in the target. This sophisticated computer code including arbitrary target geometrices has been applied to the activation of spacecraft material.[4,5]

In comparison with measured cross-sections for various reactions including spallation, the results of this code or of similar codes usually agree within a factor of two. However, its use in this study was negated by it being currently unavailable at MSFC or other local facilities.

The second method, which is used in this report, is the straight-forward technique of using a modification of Eq. (1) with known cross-sections. In doing the activation calculations, Eq. (1) must account for the degradation of energy as the protons pass through the sample. For an incident flux $\phi(E)$ the reaction cross-section "seen" by each proton at position z in the target where it has energy

$$E' = E + \int_0^z \left(-\frac{dE}{dz}\right) dz \qquad (2)$$

is a composite of the cross-sections at the mean energy of the proton in each segment Δz. To account for this, we modify Eq. (1) by using

$$\sigma(E) = \sum_{i=1}^{N} \sigma(E_i'; E, z_i)\, \Delta z_i \qquad (3)$$

where E_i' is the energy at Z_i for protons of incident energy E, and the sumation over N covers the range of the particle in the sample or the effective thickness of the target. Also, Eq. (1) must be summed over the total energy spectrum of the particle flux. Thus,

$$P = \int P(E)\,dE$$

$$= \frac{N_A}{m}\frac{A}{(NPHI)(NTH)} \sum_{l=1}^{NPHI}\sum_{j=1}^{NTH}\sum_{k=1}^{NE} \Delta E_k \phi(E_k) \times$$

$$\sum \sigma(E'; E_k \Theta_j \phi_l Z_i)\,\Delta Z_i \qquad (4)$$

where P is the number of nuclei produced per unit flux time, $\phi(E)$ is the differential flux at energy E, $\sum_i \Delta Z_i$ sums over the small increments of effective target thickness, $\sum_k$ sums over the energy region under consideration, and $\sum_l 1/NPHI \times \sum_j 1/NTH$ account for the omnidirectional character of the proton flux. This equation does not account for secondary activation, for absorption of the protons by all inelastic processes draining the proton flux, or for edge effects caused by the finite width and length of the sample.

Secondary activation can also be calculated in a similar fashion using Eq. (4) or a modification of it. The flux of secondary particles, excluding spallation, show a characteristic evaporation peaking at low energy with direct processes resulting in the addition of line spectra at forward angles and at high energy on the tail of the evaporation spectrum. [6,7] For charged particles, this low-energy peaked distribution will probably not add significantly to the activation. However, secondary neutrons should yield activation of an (n, γ) nature; especially, if moderating material is available. Since secondary neutrons will dominate high-energy Earth-albedo neutrons and since the LDEF samples will give a measure of these neutrons, further study of the characteristics of secondary neutrons and possibly calculation should be undertaken (See Recommendations).

Calculational Techniques

A. Range and Stopping Power

The range and the stopping power for incident charged nucleons can be calculated using the empirical relations reported by Zaiden:

$$-\frac{dE}{dx} = C E^{-3/4}\left[1 - \exp(-AE)\right] \qquad (5)$$

$$R = .585 C^{-1} E^{7/4}\left(1 - \exp\left(-\tfrac{3}{7}AE\right)\right) \qquad (6)$$

where -dE/dx is the stopping power in MeV/g/cm^2 and R is the range in g/cm^2. Zaidens claims that these equations are accurate to 5 percent but a comparison to range an stopping power graphs [9] indicates the accuracy is about 2 percent in the region of interests; 1 to 200 MeV. Therefore, these equations are used in the activation calculations of this report using the A and C parameters given by Zaidens.[8]

B. Differential trapped Proton Flux

The differential flux of the Trapped Proton for a 300 nm (560 km) 30 degree orbit were supplied by John Watts [10]. Since this is the approximate orbit planned for LDEF, they are used in the activation calculations. Figure (1)* shows a graph of this flux indicating a rapid drop-off up to 1 MeV, leveling off from 1 to about 6 MeV, and an exponential decline above 6 MeV. In the computer calculations, a semilog interpolation was used at energies intermediate to the tabulated values.

C. Geometrical Considerations

The omnidirectional character of the incident protons and the "edge-effects" on the samples should be taken into account in the activation calculations. Both factors are small at low energy increasing in importants at high energy. While the "edge-effects" were considered to be negligible in the present calculations, the omnidirectional effects were known to be significant and were included in these preliminary calculations. They were included by averaging the activation produced at several angles from normal incidence to 90°. The large computer time required for these calculations limited any detailed calculations.

D. Saturation and Decay Factors

While in orbit, the activation samples do not increase in activity linearly but according to the saturation factor. After their return from orbit, the activity dies off exponentially. Thus, the activity at any given time is reduced by the factor

$$(1 - \exp -\lambda T_1)\ e^{-\lambda T_2}$$

where λ is the decay constant $\left(\frac{\ln 2}{T_{1/2}}\right)$, T1 is the time of exposure and T2 is the time since the samples were removed from exposure. Since these factors depend on T1 and T2 which will not be known until the samples are studied, they have not been considered in this study.

E. Evaluation and Selection of Proton Reaction Cross-Section

Various compilations [11,12,13,14,15] of proton reaction cross-sections have been made by several authors in the U.S.

and abroad. The reliability of these cross-sections is uncertain since most were measured using stacked-foil methods, chemical separation, beta counting techniques, NaI detectors, and questionable branching ratios. In addition, the Al-27 (p,3pn) Na-24 reaction was often used as a current (proton) monitor but the cross section for this reaction was later corrected downward by nearly 30 percent. Fortunately, recent studies on [16,17,18,19] on V-57, Co-59, and Ni up to about 50 MeV have been done with relatively consistent results being obtained. These studies used stacked-foil techniques, high resolution Ge(Li) spectrometers, and current published nuclear data. In general, their quoted accuracy is 7-10 percent, including cross-section for Fe - 56 common to the three experimental groups, and all agree to within about 20 percent when normalized to common γ-ray branching ratios. Higher-energy cross-section from reference [13] can be added to the recent data when values are consistent or, when inconsistent, normalized by interpolation or extrapolation.

Natural Indium and Tantalum are more difficult. It seems that no excitation functions above 10 MeV for Indium have been done. Cross-section for (p,n) reactions have been published [20] for energies near 18 MeV and the angular and energy distribution of reactant products [6,21] have been studied. The situation for Tantalum is that it has been studied several times [22,23,20] with inconsistent results. The reactant products have also been studied [21]. The cross-sections used in this studies are selected after evaluation and when required the data are normalized.

In the cross-section tables (Table I)* the data used is that indicated reference or by as otherwise indicated reference. Normalized data is referenced and indicated by parentheses ( ). Extrapolations are indicated by [ ].

F. Pertinent Nuclear Data

Table II* contains atomic and nuclear data required to calculate proton induced activation of the LDEF samples. Nuclear half-lives, gamma-ray energies, atomic masses, and half-lives are taken from reference (9). Reaction Q values are taken from reference (24) or from reference (16) or reference (17). Stopping power and range coefficients are from reference (18) and densities are taken from reference (25).

Results and Conclusions

A computer code entitled Induced Radioactivity by Trapped Protons (IRTRAP) was developed and used to calculate the induced radioactivity expected from the proton flux given in Fig. (1). The code, in its present form, allows only for averaging of incident flux over angles from ϕ=0 to 90° and from θ=0 to 360°. No allowance for "edge-effects" or total proton-nuclear absorption is made.

The results for the production of, V-48 and Cr-51 are given in Fig. (2) in terms of fractional activation per day expected for the prominant long-lived isotopes. Although it should be remembered that "edge-effects" and nuclear absorption will reduce the theta-averaged results, the omnidirectional character of the incident flux has a significant effect both on the shape of the activation curve and on the total activation. This should be remembered when proton fluxes are to unfolded from the actual sample activation. The preliminary nature of these results is such that they should be considered as indicators of the order of magnitude of the activation and of the trend of the fractional activation.

Proton induced reaction cross-sections for Co-59 and V-51 are available from the (p,n) threshold up to or above 200 MeV. However, cross-section are not available for Indium, are poorly known for Tantalum at all energies and are poorly known for Nickel above 50 MeV. For these reasons no calculations have been done for Tantalum and Indium.

The difficulty of calculating the induced radioactivity of the LDEF samples in a simple geometry indicates the severe problems that will be encountered with complicated geometries and with sample covering materials. The most unambigious results concerning the fluxes of inducing particles will be obtained for those samples having the simplest geometry and covering material.

Recommendations

1. The development of IRTRAP should be continued so that it includes effects due to the finite dimension of the samples ("edge-effects") and the reduction of the proton flux caused by all nuclear reaction channels. A future test of the code at a high energy proton accelerator should be considered.

2. The search for and review of reaction cross-sections should be continued, as the case warrants, to improve the reliability of the cross-sections data set or to obtain previously unpublished cross-sections. If necessary, properly parameterized nuclear model calculations could yield unmeasured cross-sections or allow for the extrapolation of known cross-section.

3. A technique for determining the efficiency of the gamma-ray detector for an extended radiation source should be made. The activated samples when counted in a low-background counting system do not represent point-sources because of the almost 2π geometry they subtend to the detector. Also, since the targets are relatively thick, self-attenuation of the emitted photons must be corrected for in the analysis. It is recommended that a program to develop a calibration source be initiated as well as one to measure and model the self-attenuation effects. Initial consideration in this project should be given to uniformly depositing a known quantity of a radioisotope standard such as Eu-152 onto a 2 inch x 2 inch thin metal plate and sealing it with a thin layer of plastic. By sandwiching this source between a sufficient number of similar plates and at various distance from the detector data can be obtained for determining the efficiency and self-attenuation.

4. While a limited study of the Zaidens empirical stopping-power and range equations was done in this report a more detailed study in regards to its use in IRTRAP should be undertaken. Furthermore, the effects of proton straggling on the IRTRAP calculations should also be studied. These studies could be performed by undergraduate or graduate students under proper supervision.

5. Since one of the goals of this proposed LDEF experiment is to study the primary sources of activation, the activation of the samples by secondaries created in the sample must be understood. This requries a careful study of the energy and angular distribution of these secondaries as well as the activation produced by them. These distributions can be calculated using nuclear models and the result compared to published values. It is recommended that future plans for such a study be made. Furthrmore, some experimental reaction studies may be required to obtain either the actual cross-sections or other parameters needed for these calculations. Besides stacked-foiled techniques, in-beam techniques may be needed for reactions not leading to unstable long-lived isotopes.

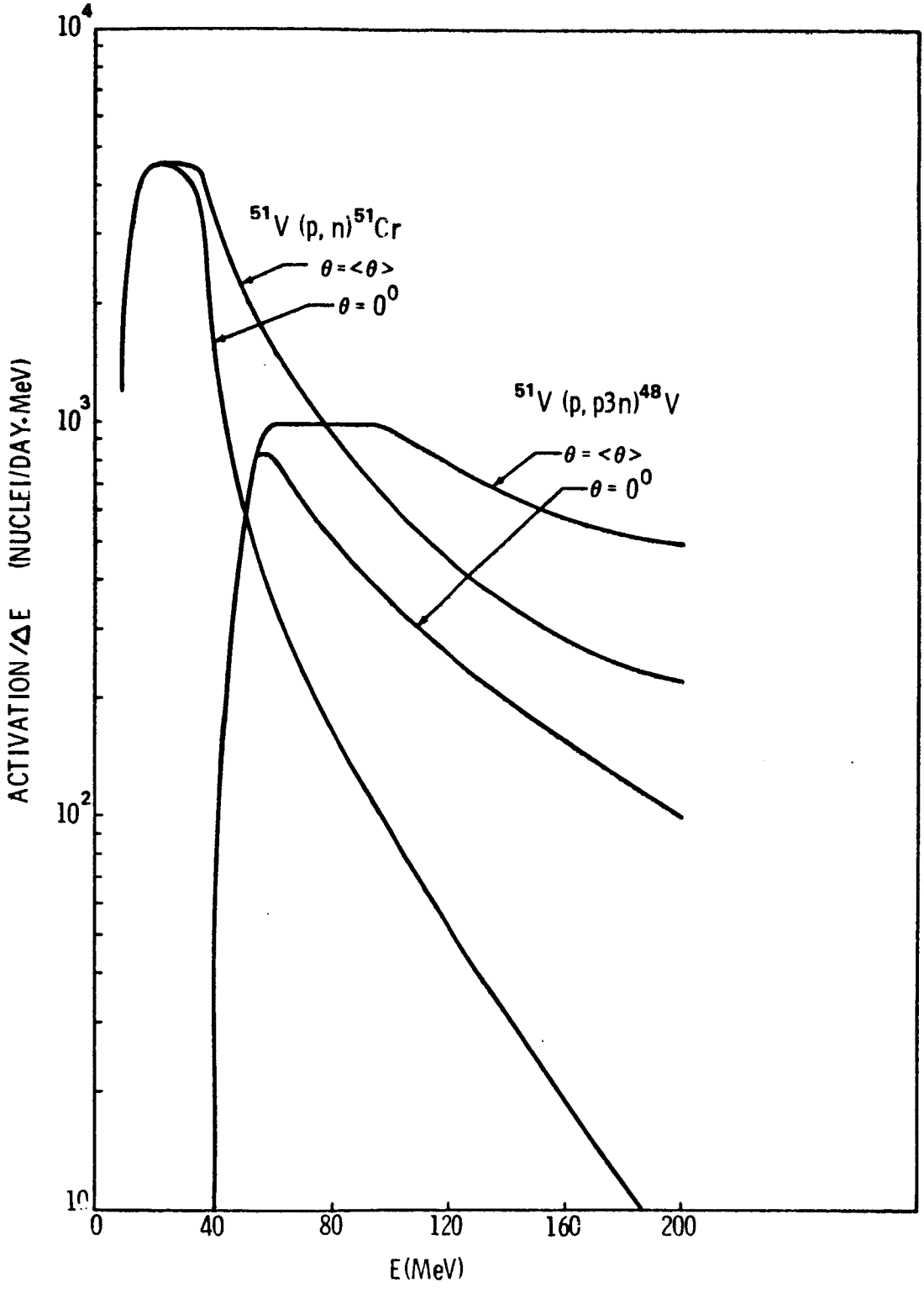

Figure 2. Fractional Activation of Vanadium

Table III: Results of Activation Calculations
Q < E < 200 MeV

Target	Isotope	Nuclei/Day Normal Incidence	Nuclei/Day Theta Average
V-51	Cr-51	143700	244900
	Cr-48	1706	3928
	V-48	52450	117700
	Sc-47	11960	26040
	Sc-46	18290	39900
Co-59	Ni-57	9742	18240
	Ni-56	399	777
	Co-58	547800	1007000
	Co-57	340000	639100
	Co-56	97840	191600
	Co-51	41430	799990
	Mn-54	82060	160000
Nickel	Ni-56	22090	42240
	Ni-57	225000	418000
	Co-55	50620	95070
	Co-56	316000	603500
	Co-57	348900	635000
	Co-58	114000	200000

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* Figure (1), Tables I and II, and other information are available upon request to the author.

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