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Radiation Damage and Activation from Proton Irradiation of Advanced Scintillators

Peter F. Bloser, Mark L. McConnell, John R. Macri, Paul J. Bruillard, James M. Ryan, Wojtek Hajdas

Abstract—We present results from a proton accelerator beam test to measure radiation damage and activation in advanced scintillator materials. Samples of $\text{LaBr}_3:\text{Ce}$ and $\text{LaCl}_3:\text{Ce}$ were exposed to protons from 40-250 MeV at the Proton Irradiation Facility of the Paul Scherrer Institute in Switzerland. Twelve energy bands were used to simulate the spectrum of the South Atlantic Anomaly (SAA), with different samples exposed to the equivalent of 4 months, 1 year, and 5 years of SAA passage. No significant decrease in light output was found due to radiation damage, indicating that these new scintillator materials are radiation tolerant. High-resolution spectra of the samples were obtained before and after irradiation with a Germanium spectrometer to study activation. We present a detailed analysis of these spectra and a discussion of the suitability of these scintillator materials for detectors in future space missions.

I. INTRODUCTION

Newly-developed advanced scintillator materials, such as the cerium-doped lanthanum halides ($\text{LaBr}_3:\text{Ce}$, $\text{LaCl}_3:\text{Ce}$), show great promise for gamma-ray detectors in future space-based astrophysics missions [1]–[3]. Their high density, high atomic number, high light output, fast decay times, and good linearity provide excellent stopping power, energy resolution, and timing properties [4]. Such detectors are potentially competitive in spectroscopy performance with solid-state detectors such as CZT, with the considerable advantages of lower material cost and well-understood, simplified readout using standard photomultiplier tube (PMT) technology.

High-energy astrophysics missions must be carried out in space, where detectors are subjected to intense photon and particle background radiation. It is thus important to verify that a given detector material is not overly sensitive to radiation damage, and that any activation of the material does not produce unacceptable levels of background. One method of checking this is to expose samples of the detector material to radiation from particle accelerators in doses equivalent to what is expected in space. In this paper we report on irradiation tests designed to mimic the proton exposure expected for an instrument in a low Earth orbit (LEO). The primary particle background in this environment is due to trapped protons in the Earth's radiation belts, particularly in the South Atlantic Anomaly (SAA).

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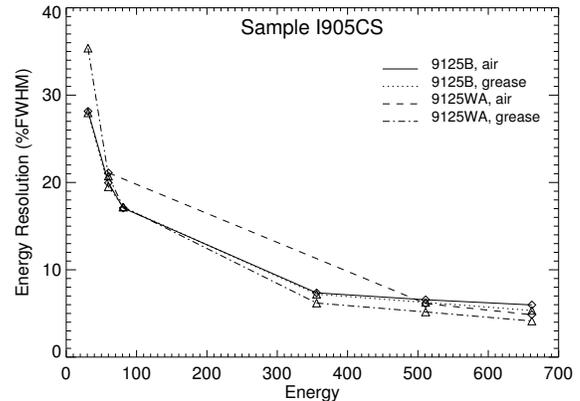


Fig. 1. Energy resolution as a function of energy for a Saint-Gobain sample of LaBr_3 . The resolution depends slightly on the optical coupling and on the type of glass in the PMT.

II. SCINTILLATOR SAMPLES

We acquired small ($0.5'' \times 0.5''$ cylinder) samples of scintillator material for exposure to proton irradiation. These included six samples of $\text{BrilLanCe}^{\text{TM}}380$ ($\text{LaBr}_3:\text{Ce}$) and six samples of $\text{BrilLanCe}^{\text{TM}}350$ ($\text{LaCl}_3:\text{Ce}$) from Saint-Gobain Crystals (5% Ce concentration). The samples were hermetically sealed in aluminum a quartz optical window. They did not include an internal optical coupling between the crystal and optical window in order to reduce the amount of passive material exposed to the proton irradiation. In addition, Radiation Monitoring Devices (RMD), Inc. provided additional samples of LaBr_3 and LaCl_3 material in the form of small chips immersed in oil and contained in small plastic vials.

The Saint-Gobain samples were characterized in the laboratory using radioactive gamma-ray line sources. The energy resolution was measured as a function of energy using two different photomultiplier tubes (PMTs) with both an air coupling and an optical grease coupling (Fig. 1). The comparison of the two PMTs was motivated by the fact that the energy resolution quoted in the Saint-Gobain data sheet that accompanied the samples was obtained using a PMT with UV-transmitting glass (the Hamamatsu R4017). We found that using a UV-sensitive PMT (Electron Tubes 9125WA) resulted in slightly better energy resolution than using a PMT with standard borosilicate glass (9125B): 4.1% FWHM vs. 5.3% FWHM at 662 keV. (We note that the energy resolution obtained with these samples is slightly worse than that normally quoted in the literature for

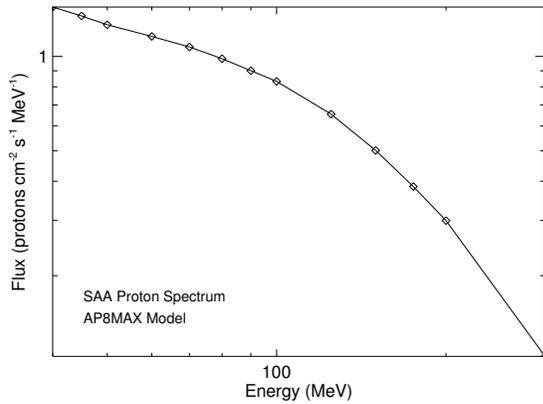


Fig. 2. Average differential proton flux spectrum in the SAA calculated using the AP8MAX model and the SPENVIS package.

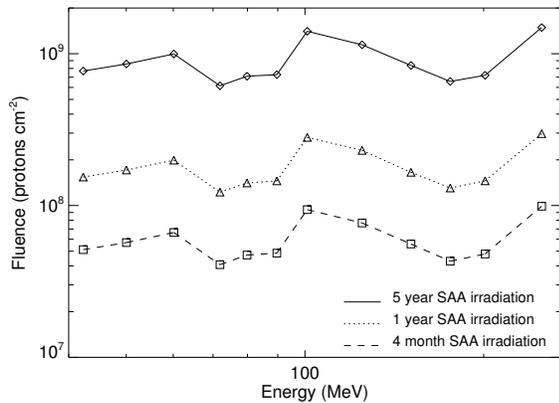


Fig. 3. Proton fluence spectra generated at PIF. The samples were exposed in three batches to represent the total fluence expected from the SAA after 4 months, 1 year, and 5 years in orbit. Note that the bin size increases at higher energies.

LaBr₃, $\sim 2.7\%$ at 662 keV, due to the lack of an internal optical coupling.) We are not certain why this should be, as LaBr₃ has not been reported to have significant scintillation light emission at UV wavelengths.

III. PROTON IRRADIATION TEST

We exposed the scintillator samples to proton irradiation at the Proton Irradiation Facility (PIF) at the Paul Scherrer Institute (PSI) in Villigen, Switzerland on 18 December 2005. Protons were produced in 12 energy bands between 40 MeV and 250 MeV to simulate the average SAA spectrum, which was calculate using the AP8MAX model and the SPENVIS package from ESA [5] (Fig. 2). The total fluence in each energy band is shown in Fig. 3; note that the bin size increases at higher energy, making the spectrum look flatter than the differential flux spectrum in Fig. 2. The samples were exposed in three batches to represent the total SAA exposure after 4 months, 1 year, and 5 years in orbit. The total (40-250 MeV) fluence was 7.3×10^8 protons cm⁻² for the 4-month exposure, 2.2×10^9

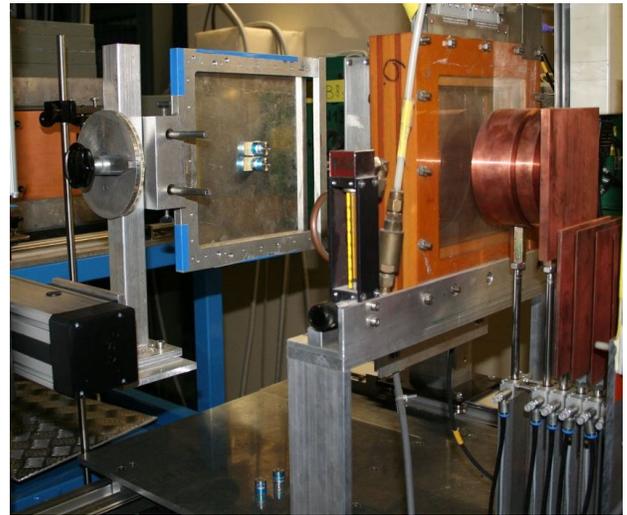


Fig. 4. Sample frame at PIF, showing irradiated samples and control samples (at bottom).

protons cm⁻² for the 1-year exposure, and 1.1×10^{10} protons cm⁻² for the 5-year exposure.

Each batch of Saint-Gobain samples comprised one LaBr₃ and one LaCl₃ crystal in the proton beam (~ 8 cm beam size) and one of each crystal placed nearby but out of the beam (the “controls”). The RMD samples were exposed in separate batches, but there were not enough of these available to use controls. The beam sample frame, with Saint-Gobain irradiated samples and controls, is shown in Fig. 4.

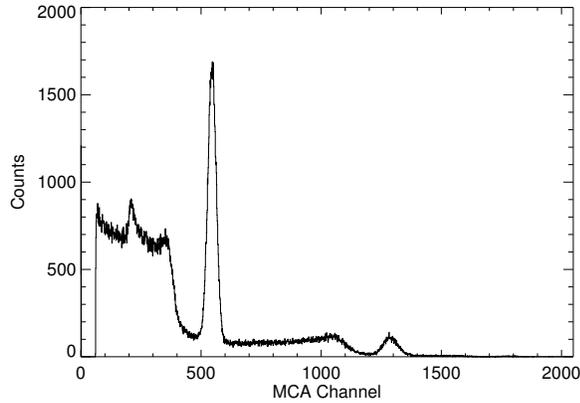
After the irradiation, two sets of measurements were made for the irradiated samples and the controls, both repeated several times over the following 24 hours: to study possible radiation damage and internal background due to activation, spectra from a ²²Na source were recorded for each detector using a PMT and multi-channel analyzer. To study the gamma-ray lines produced by the proton activation, the samples were placed in front of a calibrated high-purity Ge spectrometer.

IV. RESULTS

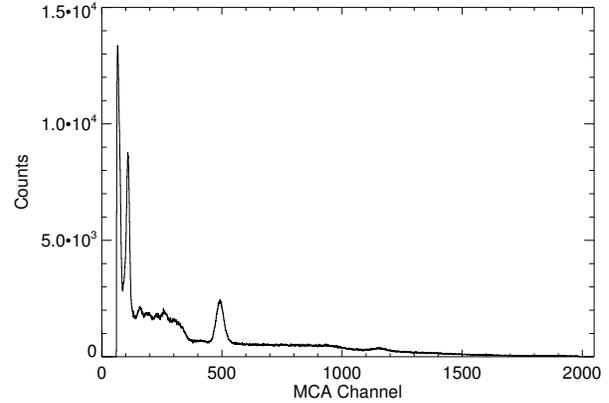
A. Radiation Damage

We looked for radiation damage by comparing the photoelectron (p.e.) yield of each sample before and after irradiation [6]. We recorded a ²²Na spectrum in each of the samples, which were placed on the entrance window of a 1.5” PMT. The before and after spectra of the 5-year-irradiated LaBr₃ sample are shown in Figs. 5(a) and 5(b), respectively. The two lines (511 keV and 1275 keV) of ²²Na were used to calibrate the spectrum and subtract any offset, and the centroid (C) and width (σ) of the 511 keV photopeak was calculated. Then the p.e. yield (p.e. keV⁻¹) is given, to within a factor of order unity, by $(C/\sigma)^2/511$.

The calculated p.e. yield of the 5-year-irradiated Saint-Gobain LaBr₃ sample is shown in Fig. 6, along with its control sample. There is evidence of slightly reduced yield following the irradiation. In Fig. 7, however, we show the p.e. yield of



(a) Before irradiation.



(b) After irradiation.

Fig. 5. ^{22}Na spectra recorded in a LaBr_3 sample before (a) and after (b) exposure to the equivalent proton fluence of 5 years of SAA passages. The bright lines at low energy in (b) are due to activation. The 511 keV and 1275 keV photopeaks from ^{22}Na are visible in both spectra.

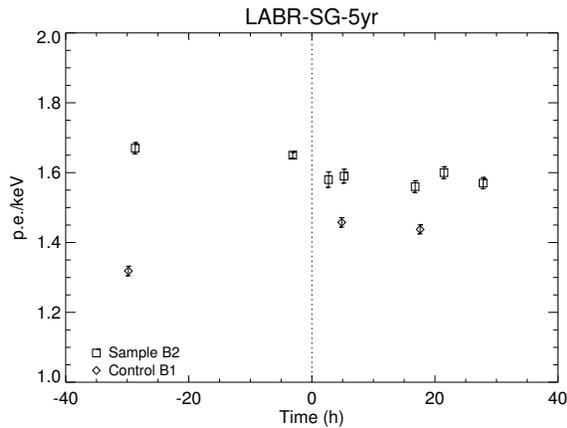


Fig. 6. Photoelectron yield of Saint-Gobain LaBr_3 sample exposed to 5-year SAA irradiation. Control sample is also plotted.

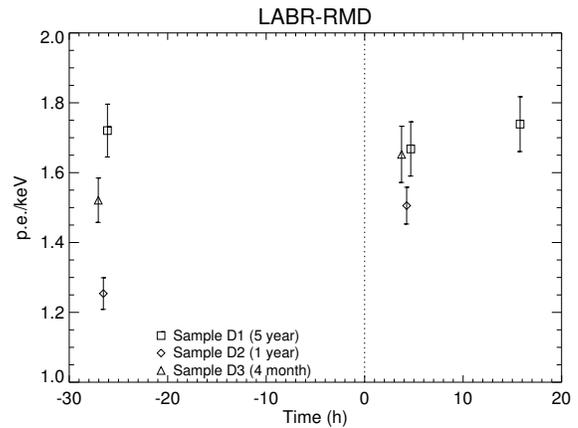


Fig. 7. Photoelectron yield of all three RMD LaBr_3 samples.

the RMD LaBr_3 samples, which are contained in a different packaging. These do not show any evidence of reduced p.e. yield. In both figures the error bars are purely statistical. We have found that there is an additional $\sim 3\%$ systematic variation in the p.e. yield from a given sample when it is removed from and replaced on the PMT several times in succession. The apparent increase in sample D2 in Fig. 7 is likely due to that sample's irregular shape, which makes it especially difficult to reliably record the same light yield in successive measurements. Other samples do not show a clear trend in p.e. yield with time, before or after the irradiation. We believe that the decrease in p.e. yield in Fig. 6 is likely due to a darkening of the quartz window and not to damage to the crystal. We conclude that there is no evidence of significant radiation damage in LaBr_3 or LaCl_3 due to proton irradiation.

B. Activation Lines

In Fig. 8 we show the Ge spectrum of the 5-year-irradiated Saint-Gobain LaBr_3 sample obtained 1.2 hours after irradiation. The fitting and identification of the lines was performed at PSI. Lines observed include ^{130}La , ^{131}La , ^{132}La , ^{132}Ce , ^{135}Ce , ^{75}Br , ^{77}Br , ^{77}Kr , ^{73}Se and ^{76}As (and ^{54}Mn and ^{24}Na from the Al housing). These are all fairly short-lived species, with half-lives of 1-2 days or less.

In Fig. 9 we show the Ge spectrum of the 5-year-irradiated Saint-Gobain LaCl_3 sample obtained 1.3 hours after irradiation. The observed lines include ^{131}La , ^{132}La , ^{132}Ce , ^{133}Ce , ^{135}Ce , ^{127}Cs and ^{129}Cs (and ^{24}Na from Al housing). The LaCl_3 shows essentially the same lines, minus the Br species, as expected. The total count rate is lower than for the LaBr_3 sample.

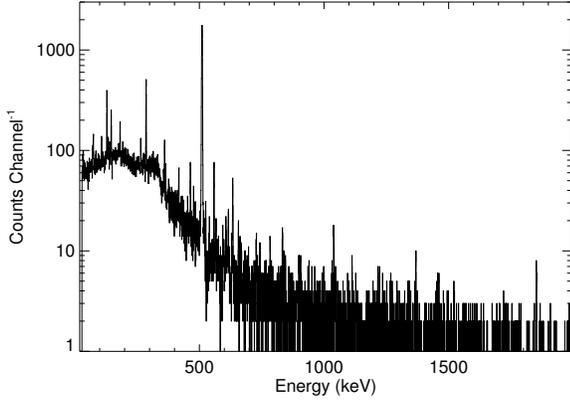


Fig. 8. Ge spectrum of activation lines from Saint-Gobain LaBr₃, 1.2 hours after irradiation with 5-year SAA proton fluence.

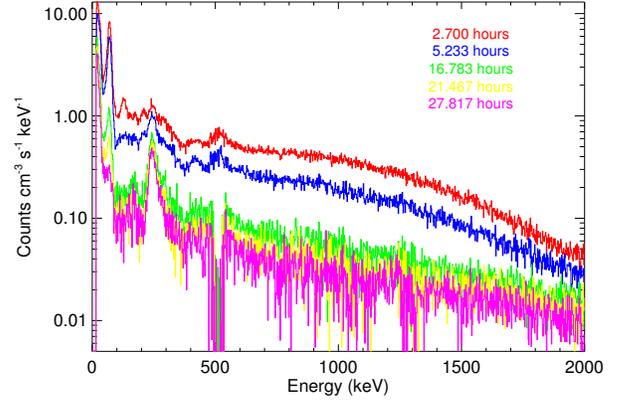


Fig. 10. Internal background due to activation in the 5-year-irradiated Saint-Gobain LaBr₃ sample at various times after irradiation.

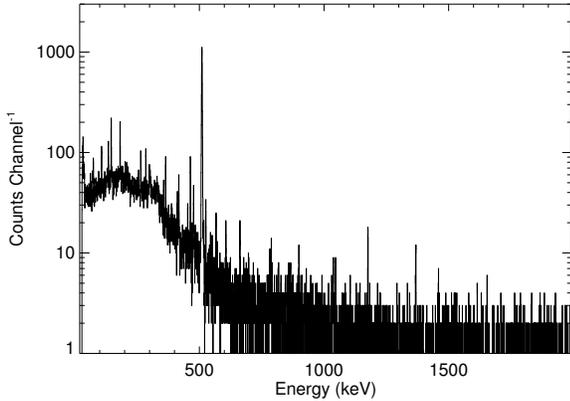


Fig. 9. Ge spectrum of activation lines from Saint-Gobain LaCl₃, 1.3 hours after irradiation with 5-year SAA proton fluence.

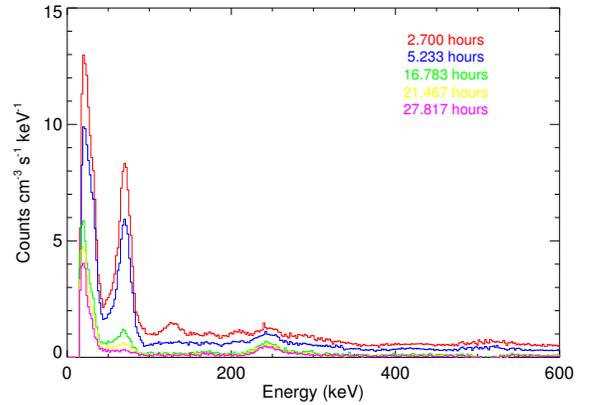


Fig. 11. Internal background lines below 600 keV due to activation in the 5-year-irradiated Saint-Gobain LaBr₃ sample.

C. Internal Activation Background

To study the internal background due to activation we subtracted the pre-irradiation ²²Na spectrum (e.g., Fig. 5(a)) from the post-irradiation spectra (e.g., Fig. 5(b)) for each sample, after calibrating and rebinning each spectrum in energy. The resulting internal background spectra for the 5-year-irradiated Saint-Gobain LaBr₃ sample is shown in Fig. 10. A beta-decay continuum is evident, as well as several lines that decay rapidly. In Fig. 11 we focus on the lines below 600 keV. The longest-lasting line is around 240 keV, probably due mainly to ⁷⁷Br. There is also evidence of ⁷⁷Kr at ~ 130 keV. There is also a bright line at ~ 70 keV that decays away with a half life of about 5 hours. The origin of this line is not clear. An important clue is that it also appears in the control sample, albeit much more weakly, and we therefore conclude that it must be caused by neutron activation. Monte Carlo simulations of the proton beam setup predict a flux of ~ 10⁻⁴ neutrons cm⁻² proton⁻¹ above 1 MeV at the sample frame, and so the 5-year irradiated samples should have received ~ 10⁵ neutrons cm⁻². The only isotope that we have been able to identify at

the time of this writing that fits the data is ^{111m}Pd, which has a half life of 5.5 hours, lines at 70.4 keV and 172.2 keV (there is evidence of both in Fig. 11), and is produced by fast and thermal neutron activation. There is no reason to expect this isotope to be present, however.

The internal background due to activation in the 5-year-irradiated Saint-Gobain LaCl₃ sample is shown in Figs. 12 and 13. Continuum and line emission is seen in this sample as well. We note that the energy resolution in this sample is poorer than that in the LaBr₃ sample shown in Figs. 10 and 11. The longest-lasting line is at ~ 250 keV, probably due to ^{137m}Ce (which must contribute to the line near 240 keV in Fig. 11 as well). Notably, there is no line evident at 70 keV. This makes the identification of the isotope responsible for the 70 keV line in the LaBr₃ samples even more problematic.

The total (15-2000 keV) internal counting rate due to activation is shown for LaBr₃ in Fig. 14 and for LaCl₃ in Fig. 15. For both LaBr₃ and LaCl₃ the counting rate decays on a time scale of about 10-15 hours. A satellite in LEO would pass through the SAA on 2-3 consecutive orbits, and then would miss it for most

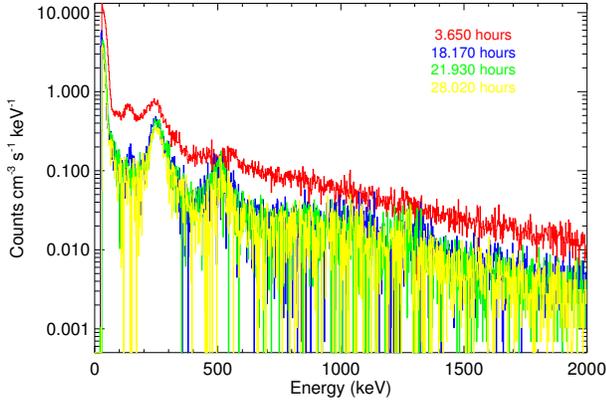


Fig. 12. Internal background due to activation in the 5-year-irradiated Saint-Gobain LaCl_3 sample at various times after irradiation.

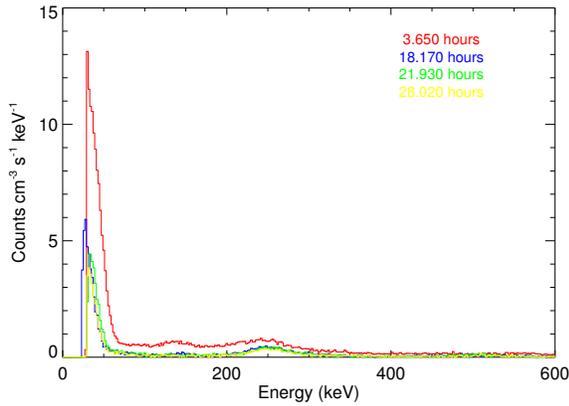


Fig. 13. Internal background lines below 600 keV due to activation in the 5-year-irradiated Saint-Gobain LaCl_3 sample.

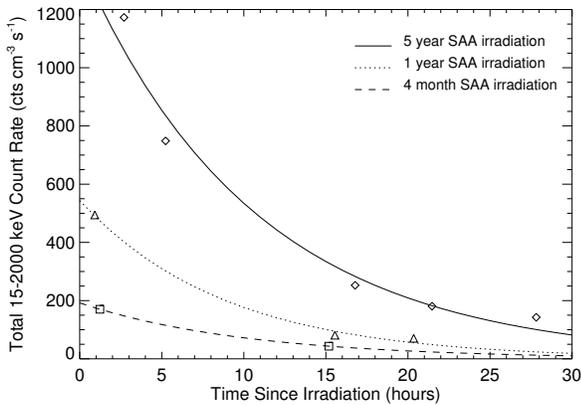


Fig. 14. Total internal counting rate (15-2000 keV) due to activation in LaBr_3 for three different proton fluences.

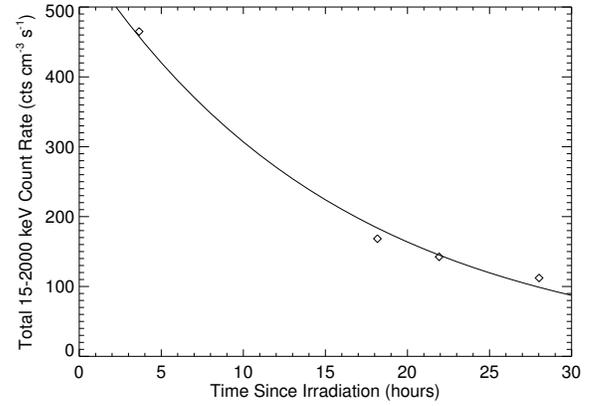


Fig. 15. Total internal counting rate (15-2000 keV) due to activation in LaCl_3 for a proton fluence equivalent to 5 years of SAA passages.

of a day. Scaling from Figs. 14 and 15, the internal background due to several hours of SAA exposure would be $\ll 10 \text{ cts cm}^{-3} \text{ s}^{-1}$, and this would substantially decay away before the next set of passages. We therefore conclude that proton activation should not be a dominant source of background for an instrument in LEO employing LaBr_3 or LaCl_3 detectors.

V. CONCLUSION

We have found that LaBr_3 and LaCl_3 do not suffer significant radiation damage after proton irradiation equivalent to 5 years in LEO, and that activation is short-lived and should not be a dominant source of background. We note that similar results have recently been reported elsewhere [7]. Our next task will be to understand the activation in detail using Monte Carlo modeling. Specifically, we plan to use MGGPOD [8], [9], a simulation suite designed to model gamma-ray background in space missions, to reproduce the results of the proton irradiation test. With our simulation tools validated, we will be in a position to confidently predict the sensitivity of specific future gamma-ray astronomy missions based on advanced scintillators.

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