Photoneutron Detection in Lightning by Gadolinium Orthosilicate Scintillators

Y. Wada, K. Nakazawa, T. Enoto, Y. Furuta, T. Yuasa, K. Makishima, and H. Tsuchiya

I. INTRODUCTION

Since the first detection reported by Shah et al.[1], thunderstorms and lightning discharges have been thought to have an ability to produce neutrons in the atmosphere[2–10]. At first, neutrons were considered to be produced via deuteron-deuteron fusion[2](H(2H,n)3He) of vapor molecules in hot lightning paths[1, 7, 11]. On the other hand, the discovery of high-energy phenomena in the atmosphere such as terrestrial gamma-ray flashes (TGFs) have convinced that thunderstorms can produce neutrons via photonuclear reactions with gadolinium orthosilicate scintillation crystals installed at sea level. Two gadolinium isotopes included in the scintillation crystals, 155Gd and 157Gd, have large cross sections of neutron captures to thermal neutrons such as 155Gd(n,γ)156Gd and 157Gd(n,γ)158Gd. De-excitation gamma rays from 156Gd and 158Gd are self-absorbed in the scintillation crystals, and make spectral-line features which can be distinguished from other non-neutron signals. The neutron burst lasted for ~100 ms, and neutron fluences are estimated to be >52 and >31 neutrons cm–2 at two observation points inside the power plant. Gadolinium orthosilicate scintillators work as valid detectors for thermal neutrons in lightning.

During a winter thunderstorm on November 24, 2017, a downward terrestrial gamma-ray flash took place and triggered photonuclear reactions with atmospheric nitrogen and oxygen nuclei, coincident with a lightning discharge at the Kashiwazaki-Kariwa nuclear power station in Japan. We directly detected neutrons produced by the photonuclear reactions with gadolinium orthosilicate scintillation crystals installed at sea level. Two gadolinium isotopes included in the scintillation crystals, 155Gd and 157Gd, have large cross sections of neutron captures to thermal neutrons such as 155Gd(n,γ)156Gd and 157Gd(n,γ)158Gd. De-excitation gamma rays from 156Gd and 158Gd are self-absorbed in the scintillation crystals, and make spectral-line features which can be distinguished from other non-neutron signals. The neutron burst lasted for ~100 ms, and neutron fluences are estimated to be >52 and >31 neutrons cm–2 at two observation points inside the power plant. Gadolinium orthosilicate scintillators work as valid detectors for thermal neutrons in lightning.

I. INTRODUCTION

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TGFs are brief bursts of energetic photons lasting for hundreds of microseconds, coincident with lightning discharges. They have been routinely detected by in-orbit gamma-ray monitors such as Reuven Ramaty High Energy Solar Spectroscopic Imager[16], AstroRivelatore Gamma a Immagini Leggero[17], Fermi[18, 19], and Atmosphere-Space Interactions Monitor[20, 21], after the discovery by Compton Gamma-Ray Observatory[22]. Besides space-borne observations of upward-oriented TGFs, similar downward-oriented phenomena have been reported by ground-based experiments, which are now called “downward TGFs”[9, 10, 23–31]. Both upward and downward TGFs originate from bremsstrahlung of relativistic electrons accelerated and multiplied by high electric fields in lightning.

Energy spectra of TGFs were found to extend up to 40 MeV[15–17, 32]. High-energy photons of >10 MeV can trigger photonuclear reactions with atmospheric nuclei such as 14N(γ,n)13N (threshold 10.55 MeV) and 16O(γ,n)15N (15.66 MeV). Neutrons generated by photonuclear reactions have kinetic energies of MeV, and are gradually thermalized in the atmosphere via multiple elastic scatterings with 14N[9, 10, 33]. When photoneutrons are produced at a low altitude, i.e. during low-charge-center winter thunderstorms, a part of neutrons arrives at the ground while the rest is captured by ambient 14N via a neutron capture 14N(n,γ)15N or a charged-particle reaction 14N(n, p)14C[34]. Therefore, neutrons can be detected by ground-based apparatus in that case.

Detection techniques of thermal neutrons have been developed in various fields, such as astroparticle physics, nuclear security, non destructive inspection, etc. Common reactions utilized to detect thermal neutrons are neutron captures 1H(n,γ)2H, 3He(n, p)2H, 6Li(n, α)3He, and 10B(n, α)7Li. For example, proportional counters filled with BF3 (including 10B) or 3He gas detected thermal neutrons in lightning at previous studies[1–5, 7]. In addition to these reactions, gadolinium isotopes 155Gd and 157Gd have drawn attention as another neutron detection scheme[8, 37–39] thanks to their high cross sections to low-energy neutrons. Here we report a direct neutron detection by gadolinium orthosilicate scintillators coincident with a lightning discharge during a winter thunderstorm in Japan.
II. INSTRUMENT

Gadolinium orthosilicate scintillator (celium-doped Gd$_2$SiO$_5$: GSO) is a relatively new type of inorganic scintillation crystals. GSO is characterized by high density (6.7 g cm$^{-3}$), fast decaying of scintillation light (40 ns), and high radiation resistance ($>10^8$ Gy). They were employed for the Hard X-ray Detector onboard the Japanese X-ray astronomy satellite Suzaku[40, 41].

Since GSO scintillators contain gadolinium isotopes, they are thought to be suitable for thermal neutron detection[42]. The upper panel of Figure 1 shows cross sections of neutron captures with stable gadolinium isotopes. The isotopes $^{155}\text{Gd}$ (14.8% in nature) and $^{157}\text{Gd}$ (15.7%) have significantly high cross sections of neutron captures to thermal neutrons (0.025 eV) of $6.1 \times 10^4$ and $2.5 \times 10^5$ barns, respectively. As shown in the lower panel of Figure 1, a 5-mm thick GSO scintillator stops almost all neutrons below 0.3 eV via neutron captures $^{155}\text{Gd}(n,\gamma)^{156}\text{Gd}$ and $^{157}\text{Gd}(n,\gamma)^{158}\text{Gd}$. After Gd nuclei capture a neutron, they emit de-excitation gamma rays; $^{156}\text{Gd}$ mainly emits gamma-ray lines at 88.97 and 199.22 keV, and $^{158}\text{Gd}$ at 79.51 and 181.94 keV[35]. The de-excitation lines are self-absorbed in the GSO scintillators, and hence they make a clear spectral-line feature for neutron detection.

The Gamma-ray Observation of Winter Thunderclouds (GROWTH) experiment has been successfully operated in coastal areas of the Sea of Japan since 2006[10, 36, 43–48]. One of our observation sites, the Kashiwazaki-Kariwa nuclear power station of Tokyo Electric Power Company Holdings in Niigata Prefecture, Japan, was upgraded with four gamma-ray detectors in 2016. The locations of the gamma-ray detectors are shown in Figure 2. Based on the discovery of photo-neutron productions in winter lightning[9, 10], GSO scintillators were added to the four detectors in 2017 for neutron detection. We utilized GSO scintillators of $2.4 \times 2.4 \times 0.5$ cm$^3$. These are connected with a photomultiplier tube of Hamamatsu R7600U, and read out by our original data acquisition system[10, 47, 49].

III. CALIBRATION IN LABORATORY

This laboratory calibration aims at confirming spectral features of neutron captures by Gd nuclei, and constraining a conversion factor to estimate the number of neutron captures in the GSO scintillators from intensities of de-excitation lines. The intensity of de-excitation lines is affected by various processes: branching ratios of de-
excitation lines, detection efficiency of gamma-ray photons inside scintillators, and simultaneous self-absorption of multiple de-excitation lines. To take these effects into account, we performed a calibration measurement by irradiating neutrons to a GSO scintillator. We utilized $^{252}$Cf as a neutron source, which exhibits spontaneous fissions with a half life of 2.645 years; 0.188 neutrons are emitted per a decay on average. The energy spectrum of neutrons emitted from this isotope follows $E^{9.5}\exp(-E/1.656\text{ MeV})$, where $E$ is the kinetic energy of neutrons\cite{50}. The $^{252}$Cf source utilized here had a radioactivity of 30 kBq at the moment of the measurement, calibrated by the manufacturer of this source; $3.5 \times 10^3$ neutrons were emitted per second. Note that 30\% systematic uncertainty is claimed to the radioactivity.

The measurement setup is shown in Figure 3. A lead block of 5-cm thickness, a tin plate of 3-mm thickness, and a paraffin block of 5-cm thickness are placed between the GSO scintillator and the neutron source. The lead block reduces background counts in GSO by screening gamma rays from $^{252}$Cf. Neutrons penetrating the lead block are thermalized by the paraffin block, then enter the GSO as thermal or epithermal neutrons. When the lead block absorbs gamma rays, the $\text{K}\alpha$ X-ray line at 74.2 keV can be emitted. This line contaminates the energy spectrum in GSO and be mixed up with 89.0 keV gamma rays from $^{155}$Gd because its cross section to thermal neutrons is one forth of $^{157}$Gd. In the same way, the center of the $\sim$260-keV line is determined to be 258.6$\pm$1.1(stat)$\pm$10.4(sys.) keV. This line is consistent with a simultaneous detection of 79.5-keV and 181.9-keV lines from $^{158}$Gd as one line at 261.4 keV. In addition, $^{156}$Gd and $^{158}$Gd emit 38.7-keV and 29.3-keV electrons by internal conversions instead of 89.0-keV and 79.5-keV gamma rays, respectively\cite{42}. The line structure around 35 keV seems to originate from monochromatic electrons of the internal conversion.

A Monte-Carlo simulation was then performed to test the number of neutrons captured in GSO in the geometry of the present experiment. A mass model of the geometry shown in Figure 3 is implemented in the simulation. Neutrons with the spectrum from $^{252}$Cf fissions were generated isotropically, then, the number of the reactions $^{155}$Gd($n,\gamma$)$^{156}$Gd and $^{157}$Gd($n,\gamma$)$^{158}$Gd is registered. When neutrons are captured in GSO, tracking of their secondary products was terminated. Here we employed the neutron cross-section database JENDL-4.0\cite{35}, developed and distributed by Japan Atomic Energy Agency.

When $10^9$ neutrons were generated in the simulation, $2.16 \times 10^5$ reactions of $^{155}$Gd($n,\gamma$)$^{156}$Gd and $7.45 \times 10^5$ reactions of $^{157}$Gd($n,\gamma$)$^{158}$Gd were registered. For the present geometry, the ratios of the reactions $^{155}$Gd($n,\gamma$)$^{156}$Gd and $^{157}$Gd($n,\gamma$)$^{158}$Gd to the total number of the generated neutrons are 0.022\% and 0.075\% respectively, and 0.097\% in total.

Then, the simulation and the measurement are compared. The neutron source $^{252}$Cf emitted ($3.5 \pm 1.1$) $\times 10^3$ neutrons s$^{-1}$ at the moment of the calibration measurement. Combining the neutron-emission rate with the ratio 0.097\% obtained by the simulation, an expected neutron-capture rate in the GSO scintillator is $3.4 \pm 1.1$ neutrons s$^{-1}$. For comparison, the calibration measurement derived that the main 80-keV peak in Figure 4 has an intensity of 0.794$\pm$0.009 count s$^{-1}$. Therefore, one neutron-capture reaction inside a GSO scintillator makes 0.23$\pm$0.08 counts at 80-keV. We adopted this number as a conversion factor to estimate neutron fluences in the following sections.

![Figure 4: A background-subtracted spectrum of neutron captures in the GSO scintillator measured with the calibration setup.](image)
IV. OBSERVATION

At 10:03:02, November 24th, 2017 (in Coordinated Universal Time), our gamma-ray detectors and monitoring posts operated by the power station recorded a downward TGF, as we previously reported[36]. The downward TGF was followed by de-excitation gamma rays of neutron captures in the atmosphere, originating from photonuclear reactions. At the same time as the detection of the downward TGF and the photonuclear reactions, the GSO scintillators also recorded an increase in count rates lasting for ~100 ms. Count-rate histories obtained by the GSO scintillators are shown in Figure 5. Significant increases in count rates were observed by Detectors A and D coincident with the lightning discharge.

Energy spectra recorded by Detectors A and D were extracted from 10 ms to 200 ms after the lightning discharge, and presented in Figure 6. Since the initial 10 ms was disturbed by the downward TGF itself, this time domain was excluded for spectral analysis. Both spectra have a significant line feature at a low energy range around 80 keV. The center energy of the line was evaluated as $83.1 \pm 2.8$ (stat.) $\pm 3.2$ (sys.) keV and $80.7 \pm 1.9$ (stat.) $\pm 3.2$ (sys.) keV for Detectors A and D respectively by fitting with a Gaussian function plus a constant component. This is consistent with the center energy at 81 keV, obtained by the calibration measurement. Therefore, this is a successful detection of neutrons by GSO scintillators.

The photon counts at the line were also evaluated to be $71 \pm 18$ counts and $116 \pm 23$ counts for Detectors A and D respectively by the spectral fitting. By utilizing the conversion factor $0.23 \pm 0.08$ counts per one neutron capture obtained by the calibration, $(3.1 \pm 1.3) \times 10^2$ and $(5.0 \pm 2.0) \times 10^2$ neutrons were captured in the GSO scintillators of Detectors A and D, respectively.

V. DISCUSSION

The GSO scintillators employed in the present study have a detection area of $2.44 \times 2.44$ cm$^2$. For thermal neutrons, whose kinetic energies are 0.025 eV or less, the detection efficiency is almost 1.0 (Figure 1), and the effective area of the GSO scintillators is 5.76 cm$^2$. In the actual situation, however, neutrons are not totally thermalized, and epi-thermal and fast neutrons must be also reaching the ground (e.g. Figure 3 in Bowers et al.[9]), not interacting with GSO (e.g. Figure 3 in Bowers et al.[9]) Therefore, we can only estimate lower limits of neutron fluences on the ground, based on the recorded number of neutron captures; $>31$ neutrons cm$^{-2}$ and $>52$ neutrons cm$^{-2}$ for Detectors A and D, respectively.

In our previous publication[36], we estimated the height, position, and the number of avalanche electrons of the downward TGF based on the on-ground measurement of radiation doses by monitoring posts. The footprint of the downward TGF was located 100 m southwest from Detector A, as shown in Figure 2. In the present result, GSO scintillators of Detectors A and D observed a significant number of neutrons, while Detectors B and C did not. Therefore, a larger number of neutrons were generated by the downward TGF around Detectors A and D, rather than around Detectors B and C. This is consistent with our previous estimation of the footprint[36]. The height and the number of avalanche electrons of the downward TGF had been also estimated to be $2.5 \pm 0.5$ km and $8^{+1}_{-4} \times 10^{18}$ electrons (above 1 MeV)[36]. To compare the estimation and the present result of neutron fluences, we need end-to-end Monte-Carlo simulations calculating photonuclear reactions and propagation of neutrons in the atmosphere, which will be covered as a future work.

This paper presents that neutrons reaching the ground were directly detected by GSO scintillators coincident with a lightning discharge. Besides neutrons, de-
excitation gamma-ray photons via atmospheric neutron captures \(^{14}\text{N}(n, \gamma)^{15}\text{N}\) also reached the ground simultaneously. In even such a high-radiation environment, GSO scintillators work as valid detectors for thermal neutron, as de-excitation gamma-ray lines of neutron captures with gadolinium isotopes were self-absorbed and clearly identified in energy spectra.

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