Comparison of Fluorescence Properties for Single Crystal and Polycrystalline YAG:Ce


Abstract—For more than a century, materials that emit visible light when exposed to ionizing radiation, or “fluors,” have been used for a variety of scientific and engineering purposes. The term “half brightness dose” ($N_{1/2}$) was developed as a consistent figure of merit to evaluate the effectiveness of a material to emit fluorescence as a function of radiation exposure. Research indicates that certain properties, such as half brightness dose, fluorescence intensity, and prompt decay time, could depend on crystalline structure. The average 3 MeV proton $N_{1/2}$ for a polycrystalline YAG:Ce paint was found to be $1.28 \times 10^{14}$ mm$^{-2}$, which is consistent with earlier research. The 3 MeV proton $N_{1/2}$ for the virgin YAG:Ce crystal was found to be 3.1 times larger than was measured for the polycrystalline paint. Subsequent $N_{1/2}$ measurements with the crystal were slightly lower than the virgin data and larger than was obtained from the polycrystalline PPMS paint.

Index Terms—Half brightness dose, phosphors, radiation damage, YAG:Ce.

I. INTRODUCTION

For more than a century, materials that emit visible light when exposed to ionizing radiation, or “fluors,” have been used for a variety of scientific and engineering purposes. Materials should exhibit a large fluorescence efficiency in order to be used as an accelerator beam detector or positioning system. Any fluor used for this purpose could be exposed to large doses of beam radiation. A candidate fluor must be able to withstand large beam doses with a minimal reduction of fluorescence light due to irradiation.

The main goal for this research was to compare the 3 MeV proton “half brightness dose” ($N_{1/2}$) and prompt decay time for single crystal and polycrystalline yttrium aluminum garnet (Y$_3$Al$_5$O$_{12}$) doped with cerium (YAG:Ce). This information will be used to help understand fluorescence phenomena for different crystal structures.

II. HALF BRIGHTNESS DOSE

The term “half brightness dose” was coined as a consistent figure of merit to evaluate the effectiveness of a material to emit fluorescence as a function of exposure. It is defined as the amount of light required to reduce the fluorescence efficiency to one half of its original value.

In 1951, Birks and Black [1] showed experimentally that the fluorescence efficiency of anthracene bombarded by alphas varies with total dose as

$$\frac{I}{I_0} = \frac{1}{1 + \frac{N}{N_{1/2}}}$$

(1)

where $I$, $I_0$, $N$, and $N_{1/2}$ represent the fluorescence intensity, initial fluorescence intensity, total incident particle fluence, and the half brightness dose, respectively. The units of $I$ and $I_0$ are related to the number of fluorescence photons interacting with the detector. When plotting the reciprocal of the light ratio ($I_0/I$) versus proton dose, the resulting curve is linear, with the slope being equal to the inverse of the half brightness dose and the intercept being unity. The Birks and Black relation describes the deterioration of fluorescence for all materials tested over the last decade [2].

From 1989 to 2001, the authors were involved in a research program designed to measure the half brightness dose for several yttrium and gadolinium compounds at ambient temperature. Table I shows the resulting half brightness doses varies between (0.16 to 1.30) $\times 10^{14}$ mm$^{-2}$ [2]. During this research, these fluors were excited at ambient temperature using a 3 MeV proton beam from a small electrostatic accelerator.

III. SAMPLE DESCRIPTION

Two YAG:Ce crystal slices were purchased from Marketech International, Port Townsend, WA. Each slice was cylindrical in shape, 20 mm in diameter, 1 mm thick, with one side of the translucent yellow YAG:Ce polished. The slice was clipped directly to a stainless steel sample holder with the polished side facing the incident beam. The beam current was kept low to minimize charge buildup during proton irradiation. Two metal clips help conduct charge away from the front of the crystal sample.

The polycrystalline samples consist of a paint containing 70% poly (phenyl methyl) siloxane (PPMS) and 30% YAG:Ce.
The time needed to reduce the fluorescence light intensity to $e^{-1}$ (36.8%) of its original value is defined as the prompt fluorescence decay time ($\tau$). For some materials, the decay time varies as a function of temperature, which makes it useful as a sensor in hazardous environments [6].

Fig. 2 shows the fluorescence decay time as a function of temperature for the YAG:Ce crystal slice and PPMS paint samples. This data was collected at Oak Ridge National Laboratory and shows the single exponential decay time for the crystal slice is about 30% larger than was measured for the PPMS paint. Both samples show significant reductions in decay time at elevated temperatures. The ambient temperature decay time for the YAG:Ce paint is about 65 ns, which is consistent with earlier measurements [7], [8]. The measurement error for decay time in Fig. 2 is estimated to be about 5%. The corresponding lines in Fig. 2 show the best fit with temperature for both the crystal and PPMS paint data.

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IV. MEASUREMENT HARDWARE

Fig. 3 shows a diagram of the $N_{1/2}$ measurement apparatus. Protons at 3 MeV were supplied by a 5SDH-2 Pelletron accelerator at the Louisiana Accelerator Center, University of Louisiana at Lafayette. Beam from the accelerator was collimated into an oval shape with an area of $1.5 \text{ mm}^2$ with currents ranging between 38–48 nA. Pressure in the scattering chamber was less than $10^{-6}$ torr during the measurements. Current is collected on an insulated stainless steel sample holder that is attached to a linear translation feedthrough at the bottom of the scattering chamber. Samples were positioned approximately 25 mm apart on the holder.

A cylindrical aluminum Faraday cup surrounds the sample holder to collect stray electrons for a full current measurement. A hole cut into the front of the Faraday cup allows the incident beam to hit the sample. A similar hole also allows the emitted fluorescence to be externally detected. The Faraday cup is electrically connected to the holder for precise current measurement. Measured beam current is passed out of the scattering chamber using a 2.75-in Conflat flange (CFF) electrical feedthrough. This current is then converted into a potential using a 1.211 MΩ resistor connected to ground. The potential across the resistor is measured by connections to analog channel seven (ACH7) and ground (GND) on a National Instruments (NI) 2120 LabVIEW Interface. Current data is processed and saved using a Macintosh G4 computer using a NI 6024E data acquisition card.

Fluorescence is detected using a Melles Griot model 13DSI silicon photodiode with an active area of $10 \text{ mm}^2$. This detector is positioned outside a 2.75-in CFF glass vacuum window and is located about 200 mm from the sample holder. A Melles Griot model 13AMP large dynamic range amplifier converts the small currents generated in the light detector to a manageable voltage range of ±2 V. This amplifier has nine gain settings of $10 \Omega$ to $1 \text{ GΩ}$ for light-based currents of 1 pA to 100 mA, respectively. The signal amplifier is battery powered to minimize line noise effects. Signal amplifier gain was adjusted to maximize the fluorescence-induced signal without saturating the system. The output of the signal amplifier was connected to analog channel one of the NI 2120. The BNC case ground from the silicon detector and that of the amplifier were connected to the GND of the
NI 2120. Fluorescence light intensity was processed and saved by a Macintosh G4 computer with a NI 6024E data acquisition card. The NI 6024E was connected to the NI 2120 using a standard a 50-pin ribbon cable.

V. DATA ACQUISITION SYSTEM

Customized LabVIEW 6i data acquisition and analysis software was developed for the \( N_{1/2} \) measurements. The acquisition software recorded the incident beam current and the visible fluorescence output from the silicon photodiode. Both parameters are analog inputs to the acquisition computer and the data was saved in spreadsheet format. Data is collected by the LabVIEW software in time increments of greater than 0.1 s. The particle dose \( n \left( \text{ppm}^{-2} \right) \) at a given time is

\[
n = \Delta t \sum I_j = \frac{Q}{qeA} \tag{2}
\]

where \( \Delta t \) is the time increment (s), \( I_j \) is the measured beam current for the \( j \)th time increment \((A)\), \( q \) is the charge state of the incident proton \((q = 1)\), \( e \) is the electronic charge, and \( A \) is the irradiation area \( \text{mm}^2 \). The total integrated charge \( Q \) is equal to the time increment multiplied by the summation of the interval beam currents. The time increment \( (\Delta t) \) used in these measurements was 1.0 s.

The analysis software uses the data collected by the LabVIEW 6i acquisition program. It calculates \( N_{1/2} \) using the Birks and Black relation shown in (1). Variances of the data are calculated using linear least squares and related techniques.

VI. RESULTS

The 3 MeV proton \( N_{1/2} \) results for the various physical forms of YAG:Ce are shown in Table II. This data was collected at ambient temperature near the center of each sample. The average \( N_{1/2} \) for the five trials of PPMS and YAG:Ce paint was measured to be \((1.28 \pm 0.21) \times 10^{14} \text{ mm}^2\), which is consistent with an earlier measurement of \((1.3 \pm 1.1) \times 10^{14} \text{ mm}^2\) shown in Table II [2], [3]. Slight darkening of the paint surface is likely caused by the formation of free carbon knocked loose by the incident protons [9]. The five irradiated PPMS paint spots were separated by a minimum of several millimeters. No beam sputtering of the samples was observed during this research.

A plot of \( I_0/I \) versus proton dose for the virgin YAG:Ce crystal is shown in Fig. 4. When \( I_0/I = 2 \), the resulting 3 MeV proton \( N_{1/2} \) for the virgin YAG:Ce crystal was \((4.03 \pm 0.65) \times 10^{14} \text{ mm}^2\), which is 3.1 times larger than the PPMS paint.

![Fig. 3. \( N_{1/2} \) measurement apparatus pictorial diagram.](image1)

![Fig. 4. Birks and black plot for crystalline YAG:Ce.](image2)

<table>
<thead>
<tr>
<th>Sample</th>
<th>Number of ( N_{1/2} ) Trials</th>
<th>Average Net ( N_{1/2} ) (x10(^{14}) mm(^{-2}))</th>
<th>Average Current (nA)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PPMS Paint</td>
<td>5</td>
<td>1.28 ± 0.21</td>
<td>38.4 ± 5.8</td>
</tr>
<tr>
<td>Virgin crystal irradiation spot (center of sample)</td>
<td>1</td>
<td>4.03 ± 0.65</td>
<td>47.7 ± 7.7</td>
</tr>
<tr>
<td>Repeat crystal irradiation at original spot</td>
<td>1</td>
<td>1.58 ± 0.18</td>
<td>46.7 ± 5.2</td>
</tr>
<tr>
<td>Repeat crystal irradiation 8 mm below original spot</td>
<td>1</td>
<td>2.11 ± 0.46</td>
<td>46.9 ± 10.1</td>
</tr>
</tbody>
</table>
value. This measurement took 1.25 h to complete. The virgin YAG:Ce crystal emits copious amounts of green-white fluorescence during the irradiation sequence. In fact, the crystal emitted more light for the same beam area than the PPMS paint sample.

After the crystal was irradiated with protons, it was allowed to rest in the vacuum chamber for about an hour. At the end of the rest period, the holder was moved to the original irradiation position at the center of the YAG:Ce crystal. The crystal was then re-irradiated with 3 MeV protons. Not surprisingly, the resulting \( N_{1/2} \) for the re-irradiated crystal was \( (1.58 \pm 0.18) \times 10^{14} \text{ mm}^{-2} \), which is about 2.5 times smaller than the virgin sample and only slightly larger than the polycrystalline PPMS paint value.

The YAG:Ce crystal was obviously damaged by proton bombardment. In fact, small cracks near the irradiation site were observed by light microscope at the conclusion of the exposure sequence. No cracks were observed on the virgin surface before irradiation. Another measurement was completed 8 mm below the center of the YAG:Ce crystal. The resulting \( N_{1/2} \) at this point is \( (2.11 \pm 0.46) \times 10^{14} \text{ mm}^{-2} \), which is about half of the virgin crystal value. Damage caused by 3 MeV proton irradiation reduces the \( N_{1/2} \) for the YAG:Ce crystal slice to near the polycrystalline paint value. Protons damage the crystal near the surface in proportion to the total dose. As dose increases, the surface region starts to act increasingly more like a polycrystalline system, as measured in the PPMS paint. Future research will be completed to further quantify this phenomenon and will also investigate the role that temperature plays on \( N_{1/2} \) for YAG:Ce.

**VII. SUMMARY**

YAG:Ce is a unique fluor that can exist in several forms. When excited, it emits bright green-white fluorescence light centered at 525 nm with a FWHM of about 100 nm. The average 3 MeV proton \( N_{1/2} \) for the PPMS paint was measured to be \( 1.28 \times 10^{14} \text{ mm}^{-2} \), which is totally consistent with an earlier measurement. Slight darkening of the paint surface is likely caused by formation of free carbon knocked loose by the incident protons. The \( N_{1/2} \) for the virgin YAG:Ce crystal was 3.1 times larger than was measured for the PPMS paint. Subsequent \( N_{1/2} \) measurements with the crystal were much smaller than the virgin data and slightly larger than was obtained from the polycrystalline PPMS paint. Damage caused by 3 MeV proton irradiation appears to reduce the \( N_{1/2} \) for the YAG:Ce crystal slice to near the measured polycrystalline paint value. Additional research will be completed to further quantify these results.

**REFERENCES**