

Development of Fast and Dense Lu and Y mixed Oxide Ceramic Scintillators for HEP

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Abstract—Dense inorganic scintillators for future colliders are expected to have a fast scintillation decay profile to mitigate pileup at high event rates and show radiation hardness up to tens of Mrad to withstand the high flux. Lu_2O_3 is one of the densest hosts at 9.4 g/cm^3 and therefore it is attractive for HEP. Lu_2O_3 and similar binary oxides, e.g., Y_2O_3 , when doped with Yb produce a fast charge transfer transition (CTT). The luminosity of CTT vary between hosts, and we found that mixing Y with the host increases the light yield. The scintillators have been prepared using ceramic process at RMD, as their melting points are very high ($> 2,000^\circ\text{C}$). Results of optical and scintillation properties and radiation hardness studies are presented.

Index Terms— Lu_2O_3 , ceramic scintillators, ultrafast decay, radiation hard, high density

I. INTRODUCTION

Precision calorimeters made of inorganic scintillating crystals have played a crucial role in high-energy physics (HEP) discoveries [1] due to their excellent energy resolution and detection efficiency. Future HEP experiments require inorganic scintillators with ultrafast ($< 10 \text{ ns}$) decay [2,3] to handle the very high event rate. They are also expected to be extremely radiation hard, without the necessity of replacement, to withstand high integrated absorbed doses of the order of tens of Mrads. There are a few scintillation mechanisms that provide ultrafast scintillation, including the charge transfer (CT) luminescence [4]. Unlike optical transitions that take place between the well-defined levels of a single activator, CT emission involves a change in the charge state of the luminophor itself. Since there are no specific rules governing such transitions, they are fully allowed, resulting in very fast scintillation with a decay time of the order of a nano second. The Lu_2O_3 doped with Yb has been identified [4] as a very promising scintillator due to its fast charge transfer transitions (CTT). In addition, Lu_2O_3 is one of the densest hosts at 9.4 g/cm^3 , which makes it very attractive for HEP. We found that mixing Y with the host increases the light yield. The melting point of these materials are very high ($> 2,000^\circ\text{C}$) and therefore these samples are manufactured using ceramic process at RMD. We are currently developing manufacturing processes to produce both pure and Y mixed Lu oxides in large sizes. Large size samples of Lu_2O_3 doped with Yb were manufactured and fabricated at RMD. These samples

were investigated both at RMD and Caltech. Some results of our studies are described in the following sections.

II. EXPERIMENTAL DETAILS

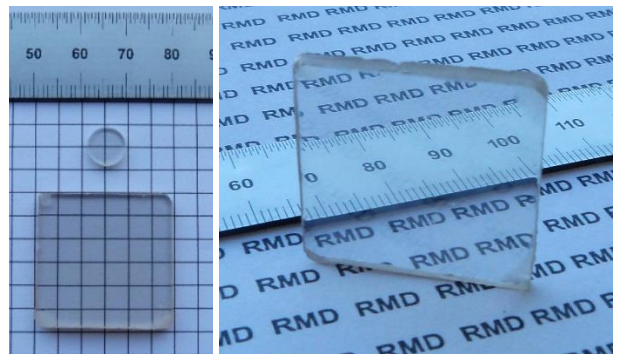


Fig. 1 (left) Small diameter disk and large size (1 inch square and 5 mm thick) samples of Lu_2O_3 doped with 1% Yb manufactured at RMD. (right) Picture of the large sample shows its excellent transparency.

For ceramic manufacturing oxide powders of high purity were obtained from different vendors or synthesized at RMD. The powders were consequently consolidated into transparent ceramics. The pictures of produced Lu_2O_3 samples doped with 1% Yb are shown in **Fig. 1**. We produced samples include small diameter (8 mm) disks and 5 mm thick plates of 1-inch square cross section. The mixed $(\text{Lu},\text{Y})_2\text{O}_3$ samples were doped with 3% Yb.

Pulse height spectra were measured with a standard nuclear instrumentation setup comprising of a preamplifier, spectroscopic amplifier, and an MCA. A super bialkali PMT (R6233-100) was used for these experiments. The light yield of each sample was estimated by comparing with a ^{137}Cs spectrum measured with a 5 mm^3 PWO crystal obtained from Epic under the same experimental settings. Decay times were measured by coupling samples to a fast PMT (R980/H6610). A 1 GHz digital oscilloscope was used to record the PMT pulses. X-ray generated emission spectra (radioluminescence) were measured with radiation from a Source-1 portable X-ray tube, with power settings of 40 kV and 0.03 mA. The scintillation light was passed through a McPherson monochromator and detected by a cooled Burle C31034 PMT.

III. RESULTS

^{137}Cs pulse height spectra measured with small samples of Lu_2O_3 and $(\text{Lu},\text{Y})_2\text{O}_3$ are shown in **Fig. 2** left. Spectrum measured with the PWO crystal under the same experimental settings is shown for comparison. The light yield of the Lu_2O_3 sample is 2.5 times that of PWO and the light yield of 50-50 Lu-Y mixed oxide is almost 6 times that of PWO. The light yield is also dependent on the Lu-Y mixing ratio. The scintillation

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time profiles measured for these samples are shown in **Fig. 2**, right. The Lu_2O_3 samples show a decay time of 1.2 ns, and the mixed sample shows two components with most of the light in <2 ns and a slow component of ~ 8 ns. The decay components and the amount of light in slow component is dependent on the Yb concentration.

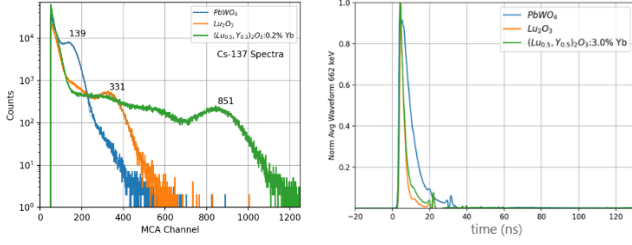


Fig 2 Comparison of Lu_2O_3 , $(\text{Lu},\text{Y})_2\text{O}_3$, and PWO (left) ^{137}Cs pulse height spectra (right) and the scintillation decay times.

Pulse height spectra measured with a ^{137}Cs source for the large plate sample is shown in top left in **Fig. 3**. A spectrum measured with the PWO crystal is also shown for comparison of light yields. We find that the light yield is consistently higher than PWO, as observed for smaller samples. This shows the scale-up to large sizes is possible without loss in light yield. The plot on top right shows ^{137}Cs and ^{22}Na spectra in linear scale. The energy resolution at 662 keV is measured to be $\sim 35\%$. The plot on **Fig. 3** bottom shows the scintillation decay profile measured with the 1-inch square plate. It shows a decay time of 1.5 ns, as seen with the small samples.

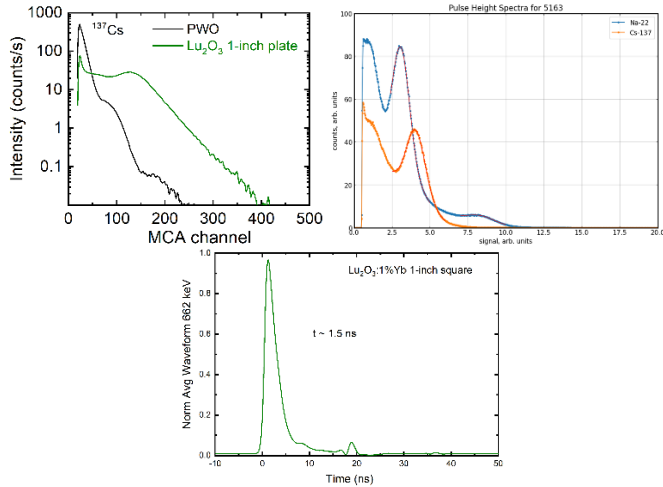


Fig 3 (top left) ^{137}Cs pulse height spectra measured for the 1-inch square sample compared with PWO (top right) ^{137}Cs and ^{22}Na pulse height spectra in linear scale (bottom) Scintillation decay profile measured with the 1-inch plate shows a decay time of 1.5 ns.

The scintillation decay times of small samples were also measured with a fast MCP-PMT at Caltech, and the measured decay profiles are shown in **Fig. 4** for 511 keV gamma-ray and ^{241}Am alpha excitations. The decay time of BaF_2 crystal was also measured for consistency. The tests show that Lu_2O_3 sample doped with 1% Yb has a scintillation decay time of 0.6 ns for 511 keV gamma-ray and 1.1 ns for the alpha particle from ^{241}Am [5]. The radiation hardness of small samples was measured at Caltech and **Fig. 5** shows the transmittance and light

output before and after irradiation up to 10 Mrad of gamma dose. The data shows that samples do not degrade after an accumulated dose of 5 Mrad and any loss of light is not significant. More results on radiation hardness will be presented for large samples.

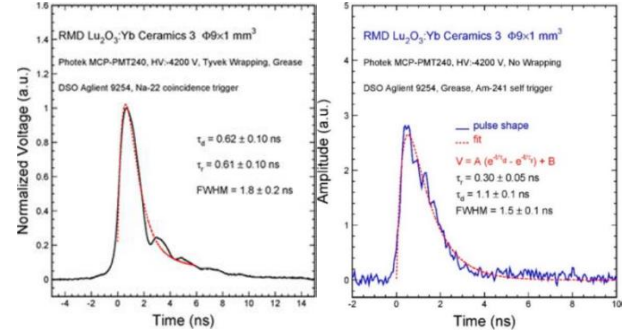


Fig 4 Scintillation decay profiles measured with a fast MCP-PMT at Caltech [5] (left) 511 keV (right) ^{241}Am alpha.

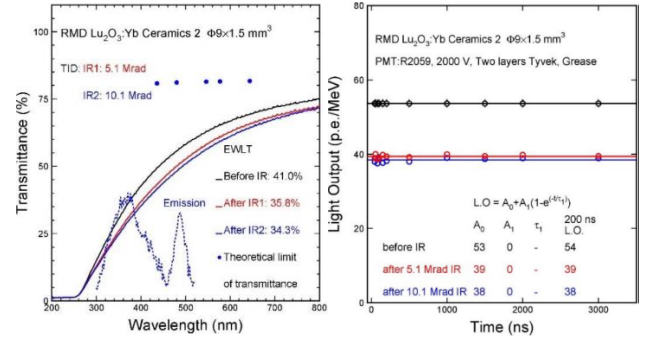


Fig 5 Radiation hardness measured with small samples at Caltech [5] (left) transmittance (right) light output before and after irradiation.

IV. SUMMARY

The results indicate the pure or mixed Lu/Y ceramic oxide scintillators doped with Yb are attractive materials for future HEP calorimetry. They show faster decay and higher light yield than PWO. The scale up to large sizes shows that transparent samples can be manufactured without loss in light. The performance of large samples is consistent with that of smaller samples, maintaining higher light yields than PWO and decay times around 1-2 ns. In addition, the emission located between 350 and 420 nm matches well with most existing photodetectors used in HEP experiments. With their high density and fast decay times the Lu-Y mixed oxides are also promising for other applications such as time of flight (TOF) or high energy radiography (in counting mode). Our future plans include further scale up to sizes as large as 20 cm in length with a cross-section of 2 cm^2 .

REFERENCES

- [1] LHCb Collaboration, "LHCb Technical Proposal, CERN/LHCC 98-4," CERN, Geneva, 1998.
- [2] P. Lecoq, A. Gektin and M. Korzhik, Inorganic Scintillators for Detector Systems: Physical Principles and Crystal Engineering, 2nd ed., : Springer, 2016
- [3] R.-Y. Zhu, "Ultrafast and Radiation Hard Inorganic Scintillators for Future HEP," *J. Phys.: Conf. Ser.*, vol. 1162, p. 012022, 2019

- [4] T. Yanagida, Y. Fujimoto, S. Kurosawa, K. Watanabe, H. Yagi, T. Yanagitani, V. Jary, Y. Futami, Y. Yokota, A. Yoshikawa, A. Uritani, T. Iguchi and M. Nikl, "Ultrafast Transparent Ceramic Scintillators Using the Yb³⁺ Charge Transfer Luminescence in RE₂O₃ Host," Applied Physics Express, vol. 4, no. 12, p. 126402, 2011.
- [5] Hu, C.; Zhang, L.; Zhu, R.-Y.; Pandian, L.S.; Wang, Y.; Glodo, J. "Novel Ultrafast Lu₂O₃:Yb Ceramics for Future HEP Applications", Instruments 2022, 6, 67