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# Development of radiation-hard scintillators and wavelength-shifting fibers

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ABSTRACT: Future circular and linear colliders as well as the Large Hadron Collider in the High-Luminosity era have been imposing unprecedented challenges on the radiation hardness of particle detectors that will be used for specific purposes e.g. forward calorimeters, beam and luminosity monitors. We perform research on the radiation-hard active media for such detectors, particularly calorimeters, in two distinct categories: quartz plates coated with thin, radiation-hard organic or inorganic compounds, and intrinsically radiation-hard scintillators. In parallel to the effort on identifying radiation-hard scintillator materials, we also perform R&D on radiation-hard wavelength shifting fibers in order to facilitate a complete active medium for detectors under harsh radiation conditions.

Here we describe the recent advances in the developments of radiation-hard scintillators and wavelength shifting fibers. We will discuss recent and projected measurements and future directions in development of radiation-hard active media.

KEYWORDS: Radiation-hard detectors; Scintillators and scintillating fibres and light guides

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#### 1 Introduction

Future circular and linear colliders as well as the Large Hadron Collider in the High-Luminosity era have been imposing unprecedented challenges on the radiation hardness of particle detectors that will be used for specific purposes e.g. forward calorimeters, beam and luminosity monitors. We performed research on the radiation-hard active media for such detectors, particularly calorimeters, in two distinct categories: quartz plates coated with thin, radiation-hard organic or inorganic compounds, and intrinsically radiation-hard scintillators. In parallel to the effort on identifying radiation-hard scintillator materials, an R&D on radiation-hard wavelength shifting fibers should also be carried out in order to facilitate a complete active medium for detectors under harsh radiation conditions. In this context, we have identified materials with proven radiation resistance, long Stokes shifts to enable long self-absorption lengths, and with decay constants ~ 10 ns or less.

Quartz Čerenkov radiators have implementations in beam and luminosity monitors as they are intrinsically radiation-hard. To improve the light production inside the quartz plates, we considered various light enhancement tools: p-Terphenyl (pTp), 4% Gallium doped Zinc Oxide (ZnO:Ga), o-Terphenyl (oTp), m-Terphenyl (mTp), and p-Quarterphenyl (pQp). After 20 MRad of proton irradiation, the light output of pTp drops to 84% of the initial level, then it slowly flattens, and after 40 MRad of radiation we still observe less than 20% loss of light production [1–3]. We have built a quartz plate calorimeter prototype consisting of 20 layers of quartz plates (15 cm × 15 cm × 5 mm, 2  $\mu$ m pTp deposited on one side) with 7 cm iron absorbers. The stochastic term of the energy resolution of this prototype for hadrons was 211 % and for electrons was 26 %. Details about these measurements can be found in [4].

We have investigated scintillators that are intrinsically radiation-hard. The samples that we probed were thin plates of Polyethylene Naphthalate (PEN) and Polyethylene Terephthalate (PET) and thin sheets of HEM. These materials have been used in beamline instrumentation but a study for calorimetry has not been performed so far. We have studied their radiation damage and recovery properties in great detail [5, 6].

Recently we developed a new scintillator material. Peroxide-cured polysiloxane bases were doped with the primary fluors p-terphenyl (pTP), p-quarterphenyl (pQP), or 2.5-Diphenyloxazole (PPO) and/or the secondary fluors 3-HF or bis-MSB. The scintillation yield of the pTP/bis-MSB

sample was compared to a BGO crystal using a setup with <sup>90</sup>Sr source and a Hamamatsu R7525-HA photomultiplier tube (PMT). The pTP/bis-MSB sample was also tested for radiation hardness and shows exciting recovery properties [6, 7].

The development of the wavelength shifting materials proceed in two directions: thin film depositions on quartz rods and quartz capillaries. For this purpose, we investigated doped ZnO:Zn/Mg, 3HF, Teflon and sylgard. Recently, we have developed Cerium-doped scintillating glasses which can also be drawn in fibers. Hence they are candidates for single solid radiation-hard wavelength shifting fibers.

Here we describe these recent advances in the developments of radiation-hard scintillators and wavelength shifting fibers and discuss past and projected measurements.

#### 2 Radiation-hard scintillators

As the very first option, quartz plates with various surface coatings were probed. Quartz is proven to be extremely radiation-hard [3], but the plates should be coated with organic and inorganic scintillators such as para-Terphenyl (pTp), Anthracene (AN) and Gallium-doped Zinc Oxide (ZnO:Ga) in order to enhance the light yield. Organic scintillators, pTp and AN, exhibit blue fluorescence under UV. The evaporation technique is used for coating pTp, and RF sputtering technique is used for coating AN on the quartz plates. The radiation hardness of pTp was extensively tested. The light yield of pTp sample dropped to 84% of the initial light yield after 20 Mrad proton irradiation, and 80% of the initial light yield after 40 Mrad proton irradiation [8]. Inorganic scintillators such as ZnO:Ga can also be coated on quartz plates to increase the light yield. ZnO:Ga has a short de-excitation time of 0.7 ns and very high luminous yield of 15k photon/MeV [9].

Also studied are intrinsically radiation-hard scintillators such as Polyethylene Naphthalate (PEN), Polyethylene Terephthalate (PET) and High Efficiency Mirror (HEM). PEN and PET are bright and inexpensive plastic scintillators. PEN was created by the Japanese company Teijin Chemicals [10]. The company measured its light yield as 10,500 photons/MeV. PEN makes intrinsic blue scintillation with an emission spectrum peak of 425 nm [11]. PET is a common type of polyster and it is widely used to make plastic bottles and as a substrate in thin film solar cells. The emission spectrum of PET peaks at 385 nm [12]. HEM is structurally a multilayer of polymer mirrors. We have made a stack of alternating slices of HEM sheet and quartz plates and tested the scintillating properties of the stack.

Figure 1 shows the response of the relevant tiles to 150 GeV muon test beam. Compared to the reference tile (denoted HE), anthracene coated quartz plates, the HEM stack and the PEN tile demonstrate promising results. Further R&D, e.g. mixtures of different components, should be performed in order to increase the efficient to detect minimum ionizing particles.

As a candidate of a novel, intrinsically radiation-hard scintillator, Peroxide-cured polysiloxane bases were doped with the primary fluors p-terphenyl (pTP), p-quarterphenyl (pQP), or 2.5-Diphenyloxazole (PPO) and/or the secondary fluors 3-HF or bis-MSB. The polysiloxane scintillator base, HARDSIL [13], and other chemicals were purchased from Gelest, Inc.. The scintillation yield of the pTP/bis-MSB sample was compared to a BGO crystal using a setup with <sup>90</sup>Sr source, and the pTP/bis-MSB was measured to yield roughly 50% better light production compared to the BGO crystal. Recently, the production process is highly optimized with upgraded custom control



**Figure 1**. The response of the HE (reference scintillator tile: SCSN-81); PEN and PET tiles; stack of HEM sheets; quartz plates with pTp and anthracene coatings to 150 GeV muon beam.

circuits and modified oven. With the recent advances, tiles of sizes from  $3 \text{ cm} \times 3 \text{ cm}$  to  $10 \text{ cm} \times 10 \text{ cm}$  can be produced as well as the so-called finger tiles of size  $2 \text{ cm} \times 10 \text{ cm}$ . All tiles can be machined to open grooves for wavelength-shifting (WLS) fibers and dimples to directly couple the Silicon Photomultipliers (SiPMs), and can be polished, with no mechanical issues.

In Summer 2017, 2 3 cm  $\times$  3 cm tiles with photodetectors (Hamamatsu S12572-15 [14]) directly coupled to dimples were tested at CERN H2 beam line [15]. One tile was polished whereas the other one was not. Figure 2 shows the pedestal charge spectrum of one of the photodetectors. The spectrum is fit to

$$f(x) = \sum_{i=0}^{N_{\text{peaks}}-1} A_i e^{-\frac{1}{2} \left(\frac{x-\mu_i}{\sigma_i}\right)^2}$$
(2.1)

where  $\mu_i = \mu_0 + iG$ . The function is basically a sum of Gaussians with independent weights  $A_i$  and widths  $\sigma_i$ ; and means  $\mu_i$  are parametrized as a function of the pedestal mean  $\mu_0$ , the peak id *i* and the SiPM gain *G* (expressed as the charge generated by a single photon). Figure 2 shows all the parameters obtained for this particular photodetector. The gain of the SiPM is a parameter of this fit function which is obtained as 48.43 fC for this particular SiPM.



Figure 2. Pedestal charge spectrum of one of the photodetectors showing a few photon peaks.

During the tests, the SiPMs couped to the dimples were downstream and the tiles were centered with respect to the beam. 150 GeV muon beam was used to measure the response of the tiles to Minimum Ionizing Particles (MIPs). The muons passing through the tile and 1 mm away from the SiPM location are selected using wire chamber profiles. Figure 3 shows the charge spectrum of the new development of scintillators in response to traversing MIPs with polished (left) and unpolished (right) configurations. The distributions are fit to Gaussian + Landau with the mean for the Gaussian and the most probable value for the Landau functions constrained to be identical, denoted as  $\mu$  in figure 3. N<sub>G</sub> and N<sub>L</sub> are the normalizations for the Gaussian and the Landau, and  $\sigma_G$  and  $\sigma_L$  are the respective widths. The mean response of the polished tile corresponds to approximately 18 photons, and of the unpolished tile approximately 14 photons. These results validate the recent production process modifications and the feasibility to extend the production to various currently unprobed specifications.



**Figure 3**. The charge spectrum of the new development of scintillators in response to traversing MIPs with polished (left) and unpolished (right) configurations.

The tiles mentioned so far were exposed to various irradiation sources and superior performances were observed. As an example, figure 4 (left) shows the radiation damage and recovery of the PEN tile after irradiation with <sup>137</sup>Cs gamma source to 14 Mrad [5]. The PEN tile recovers from an initial damage of 55 % to a permanent damage of 18 %. We have also investigated the LED stimulated recovery of the scintillators from radiation damage and have obtained promising results. Figure 3 (right) shows the effect of the red-green-blue LED stimulated recovery of the first generation polysiloxane scintilator. The LED stimulation improves the radiation damage recovery by more than 20 % [6].

#### 3 Radiation-hard wavelength shifting fibers

Previously, we validated the principle of coating quartz fibers with pTp with in-house methods. ZnO:Ga (1-4 %) [9] and CeBr<sub>3</sub> [16] are outstanding scintillators for radiation-hard wavelength shifting fiber applications. Soft WLS materials fill capillary cores via vacuum melt imbibition, such as pTp and Stilbene admixtures, bisMSB, POPOP, 3HF+naphthalene or others. We constructed demonstrative quartz capillaries with anthracene and placed a bunch of 7 fibers into an 80 GeV electron beam measuring 8–9 photoelectrons. We also performed preliminary studies of capillaries filled with 3HF [17]. The results indicate that the 3HF filled capilarries can be successfully utilized in future implementations. However, for realistic tests, larger capillaries would be needed.



Figure 4. Radiation damage and recovery of PEN (left) and LED stimulated recovery of PEN (right).

Recently, we constructed a WLS fiber as a Pt-cured silicone capillary (2.3 mm outer diameter, 1 mm inner diameter, certified for gamma sterilization) with a silicone gel conveying 3HF. Holes were drilled through the centers of three 5 mm thick blue scintillator tiles of lateral size 2.5 cm  $\times$  2.5 cm. The silicone-based WLS fiber was placed through the center hole, and it was coupled to a Hamamatsu S12572-10. The fiber was parallel to the beam direction. The response to 150 GeV muons was measured by selecting the muons passing through the tiles and 1 mm away from the SiPM/fiber using the wire chamber profiles. Figure 5 shows the charge distribution. The mean response was approximately 21 photons.



**Figure 5**. The response to traversing MIPs of the three-tile assembly read out with silicone-based WLS fiber passing through the center of the tiles and coupled to a SiPM.

Another area of investigation in terms of scintillating fibers is based on Cerium and/or Boron doped scintillating glasses.

#### 4 Conclusions

The options of intrinsically radiation-hard scintillators are limited. PEN, PET and HEM are extensively tested. The properties of their mixtures can be investigated for optimized scintillator specifications.

The recently developed polysiloxane-based scintillators enable opportunities for component tuning for optimal performance. The production process was highly optimized with upgraded custom control circuits and modified oven. The latest production tiles produce 18 photons per traversing MIP. The development is open and promising for various implementations.

Quartz is extremely radiation-hard and with the correct combination of coating and readout, it can be the optimal option for many future implementations. With the advances in microprocessing techniques, coating is a relatively easy process. Hence various other coatings as well as their mixtures can be probed for improved scintillation performance.

The achievievements on radiation-hard wavelength shifting fibers are encouraging. The latest production of silicone-based fibers are very close to be considered for real implementations.

The R&D is still underway in order to identify and study implementation-specific radiation-hard and high light yield materials.

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