Proton induced radiation damage to the fluorescence capability of plastic scintillators for the Tile Calorimeter of ATLAS

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Abstract. The Tile Calorimeter of the ATLAS detector relies on plastic scintillators to aid in the energy reconstruction of hadrons, taus, and jets of quarks and gluons that arise from the proton-proton collisions within the Large Hadron Collider of CERN. These scintillators are exposed to harsh radiation environments and therefore sustain damage. In 2018, current plastic scintillators employed in the Gap region of TileCal will be replaced with more radiation tolerant plastics. A series of investigations are currently being conducted into the radiation hardness of several polyvinyl toluene (PVT) and polystyrene (PS) based plastic scintillators which are candidates for the upgrade. In this study, we investigate the damage induced by 6 MeV protons to the light fluorescence capability of 350 μm thick scintillators. Presented here are the results of the damage for proton doses ranging between 800 kGy to 80 MGy conducted using the 6 MV tandem accelerator of iThemba LABS.

1. Introduction

Plastic scintillators are organic materials which undergo luminescence upon interaction with ionizing radiation. They are employed by the Tile Calorimeter of ATLAS, to detect the hadronic particles which result from the proton-proton collisions within the Large Hadron Collider of CERN [1]. Their properties of high light output and high optical transmission ensure that good resolution in measurements can be achieved. Their fast rise and decay times are ideal since fast timing responses are required by the detector. The main problem encountered by plastic scintillators however, is radiation damage incurred due to their interactions with the ionizing particles to be detected. This damage leads to a significant decrease in the light yield of the scintillator and may compromise the detectors performance.

With the LHC gearing up to run proton collisions at increased center of mass energy, with luminosities of a factor of 10 beyond the current design value by 2022, the radiation environment within the ATLAS detector is expected to become much harsher. The Tile Calorimeter has therefore implemented a series of upgrades in order to ensure that the detector performance can be sustained for several years to come. Part of phase two of this upgrade will be implemented in 2018 where scintillators from the GAP region of the Tile Calorimeter will be replaced with more radiation hard

plastics. To aid in the choice of a replacement candidate, a series of investigations which examine the radiation hardness of several commercially available plastic scintillators have been conducted. In this paper, results on the impact of radiation damage to the fluorescence capability of PVT and PS based plastic scintillators is presented.

2. Background

Plastic scintillators generally consist of organic fluors suspended in a polymer base. Ionizing radiation causes the molecular excitations of delocalized π-electrons typically in the base. These may de-excite through several mechanisms [2], i.e. fluorescence, phosphorescence, delayed fluorescence, radiation-less internal conversion or vibrational losses. The fluorescence process occurs for transitions from the lowest vibrational first excited state to the ground state whereby energy is emitted in the form of light of a characteristic wavelength. This wavelength ranges around 300-350 nm in general for polymer bases. The base however, tends to have a low fluorescence light yield due to the small Stoke's shift between their excitation and emission ranges.

Primary fluors are thus added in small concentrations (typically < 3% by weight). These are chosen such that their absorption spectra match the emission spectra of the base and generally contain a high quantum yield of the energy transfer transition. Light can be transferred between base and fluor via either radiative re-absorption, or by a non-radiative coulombic interaction called Forster resonance energy transfer. [3]

Forster energy transfer is limited by the distance between the interacting states and is therefore more likely to occur with increasing fluor concentrations until a saturation is reached. Light is then emitted by the fluors at higher wavelengths, generally in the UV range of 350-400 nm. Since this wavelength is still below the peak efficiency of common photomultipliers, a secondary fluor is added at concentrations of < 0.1% by weight. The secondary fluor acts as a wavelength shifter (shifting emission to 400-500 nm) and prevents re-absorption of scintillation light by the primary fluor. It also helps to increase the bulk attenuation length of the emitted light. Energy transfer between the primary and secondary fluors occurs via radiative exchange. [3]

Whilst ionizing radiation may lead to molecular excitations and hence scintillation, prolonged exposure can also cause the breaking of chemical bonds thereby modifying the polymer properties. The optical properties of the scintillator can be effected in two ways, either a decrease in actual scintillation light output due to damage to the fluorescent component, or through a degradation in its transmission character as a result of the formation of optical absorption centres. The change in transmission character further affects the light attenuation length of the scintillator.

In [4], preliminary transmission and Raman studies on 6 MeV proton irradiated PVT samples of 250 μm thickness were presented. The study looked at irradiated doses of ~1.8 kGy, 164 kGy and ~1-1.5 MGy. At these doses, an increase in transparency over the wavelength region of 300 - 410 nm was observed for the blue emitting scintillators, whilst an absorptive tint began to form for the green emitting scintillator. In [5], the study was extended to cover doses of approximately 0.8 MGy, 8 MGy, 25 MGy and 80 MGy in 350 μm thick samples where visible discolouration in samples developed. At these higher doses, the absorptive tint shifted to higher wavelengths with increasing dose and could be correlated to free radical production.

3. Methods and materials

The main properties of the scintillators under study are summarised in table 1. The light output is given relative to the light output of anthracene which has the highest known light output amongst all organic scintillators. The scintillator response times are given in terms of the rise time and decay time. The rise time characterizes the sharp increase in intensity of a measured scintillation light pulse, whilst the decay time measures the time taken for the intensity to exponentially drop to 1/e of the maximum value. The wavelength at which maximum intensity of light emission occurs is also given.

EJ208 emits at a slightly higher wavelength as compared to the other blue scintillators, whilst EJ260 emits in the green range. These higher wavelength shifting scintillators are marketed for

exhibiting greater radiation tolerance since radiation damage causes the optical attenuation of light across the lower visible wavelength range [6].

Samples cut and polished to dimensions of 5 mm by 5 mm and of 350±30 μm thickness were subjected to 6 MeV proton irradiation using the 6 MV tandem accelerator (operated at 3 MV) at the iThemba LABS, Gauteng. The procedure for irradiations has been described previously in [4] and [5]. The light fluorescence of each plastic scintillator was measured using the LabRAM HR Raman spectrograph. A 229 nm laser with a power of ~3-5 mW was employed to provide energy for molecular excitations to occur and thereby prompt light emission through luminescence. At this wavelength, the laser energy is sufficient to be absorbed by the PS or PVT base, and the successive light transfer from base to primary and secondary fluors can occur.

Table 1: Properties of the scintillators under study.

	EJ200	**EJ208**	**EJ260**	**BC408**	**UPS923A**	**TileCal**
Manufacturer:	Eljen Technology	Eljen Technology	Eljen Technology	Saint Gobain Crystals	Institute of Scintillating Materials, Kharkiv.	Institute of High Energy Physics, Protvino in association with SIA luch, Podolsk.
Base	PVT	PVT	PVT	PVT	PS	PS
Primary Fluor	0.3% organic fluors	0.3% organic fluors	0.3% organic fluors	Not available (However, listed as a performance equivalent of EJ200)	2% PTP	1.5% PTP
Secondary Fluor					0.03% POPOP	0.044% POPOP
Light Output (% Anthracene)	64	60	60	64	60	Not available
Wavelength of Max. Emission (nm)	425	435	490	425	425	
Rise Time (ns)	0,9	1	~	0,9	0,9	
Decay Time (ns)	2,1	3,3	9,2	2,1	3,3	
Source	<	[6]	>	[7]	[8]	[9]

Photographs of the LabRAM HR are shown in figure 1. The laser is guided through a series of mirrors and optics in the machine, and is incident on the sample through the Olympus microscope aperture. As the sample fluoresces, the backscattered light is passed through a 150 lines/mm grating and collected by a CCD detector to obtain a differential wavelength spectrum.

The main obstacle to overcome during testing, was the effect of photo-bleaching of the fluorescent light. For excitation wavelengths below 250 nm, photo-bleaching occurs more prominently since the probability of exciting the electron to the triplet state increases. This is a stable state with a long lifetime and can interact with other molecules to produce irreversible covalent modifications. Photo-bleaching therefore results in a decrease to the fluorescence yield since molecules undergo photon induced chemical damage.

The destruction of the molecule is proportional to the emission intensity, the emission time and the number of excitation and fluorescence cycles undergone. In order to reduce the effect of photo-bleaching undergone during testing, the laser was scanned over a 20x20 μm^2 area and the acquisition time was limited to one second per spot tested.

Three spots along the irradiated region and three spots along the un-irradiated regions of each sample were tested in order to gauge the ratio of loss to fluorescence yield over the wavelength range of 350-500 nm.

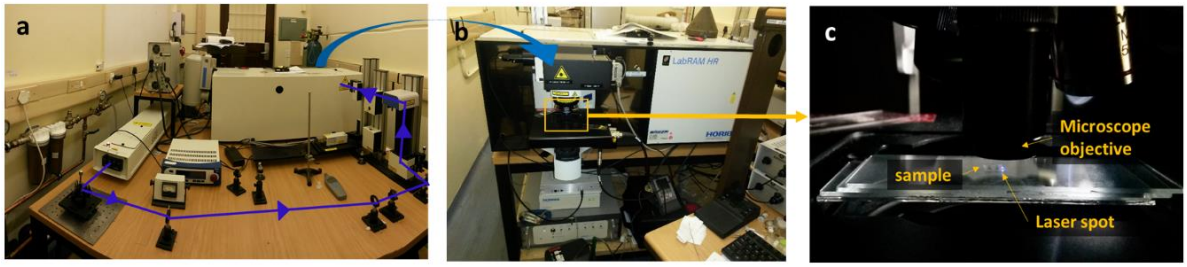

Figure 1: (a) Back view showing the path travelled by the laser into the LabRAM HR, (b) front view of the spectrograph, (c) enlarged view of the sample undergoing fluorescence

4. Results of fluorescence testing

The fluorescence spectra for EJ200 samples are shown in figure 2. An indication of the visual discolouration of the irradiated spot for the approximate dose exposures are also shown. A similar trend in the loss to fluorescence peak features were observed for the other scintillator types [10].

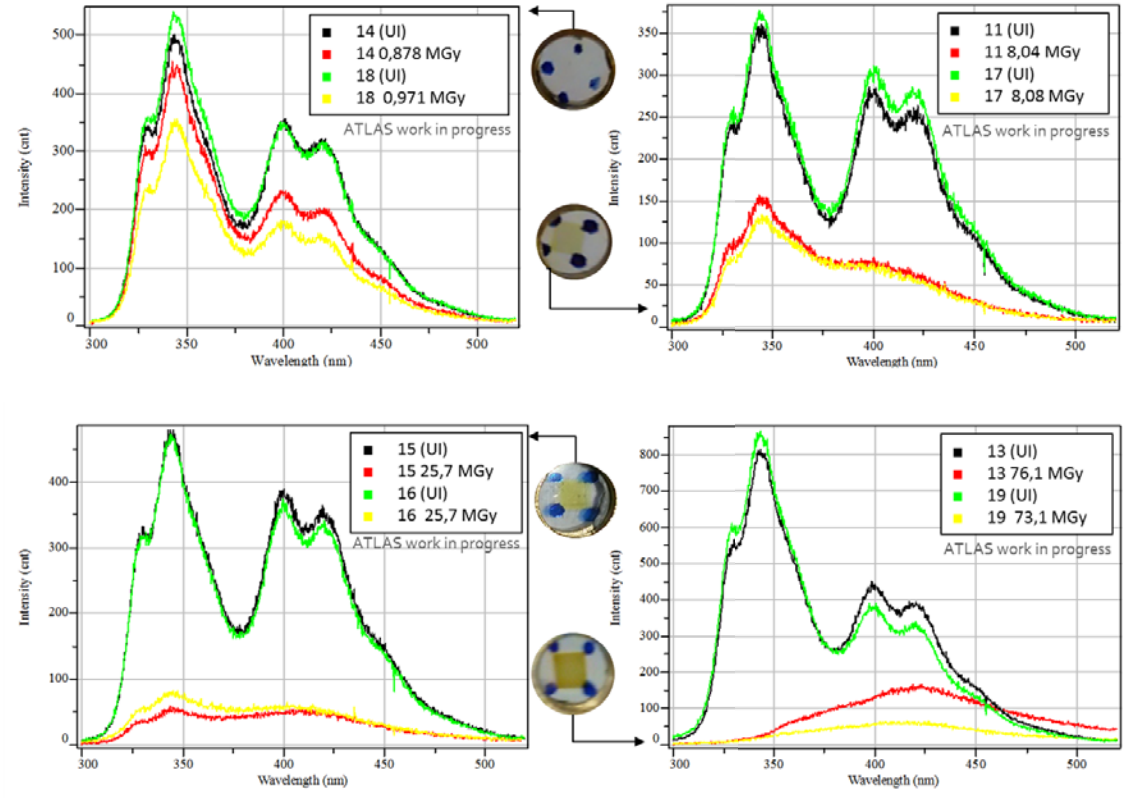

Figure 2: Fluorescence spectra of 350 µm thin EJ200 samples after exposure to several doses of 6 MeV protons

The fluorescence spectra in the un-irradiated EJ260 samples showed a very small amount of fluorescence over the 450-500 nm region despite its wavelength of maximum emission being at 490 nm. It was realized that since backscattered fluorescence light was detected, a large component of the measured light could have been from the surface which could have had less interaction with the fluors. Hence, a large component of "un-shifted" base scintillation light was measured.

For doses of ~0.8-1 MGy, an overall intensity loss occurs, with a more distinct loss to fluorescence peaks at the higher emission regions of 375-500 nm. This wavelength range correlates with emission expected from the fluor dopants. At this dose, very minimal transmission losses were observed, although a feature indicating loss to absorption by the fluors occurred. At progressively higher doses, the fluorescence intensity is further decreased, with significant loss to base emission peaks and fluor emission peaks. At the 70-90 MGy dose range, a very weak signal is obtained with an additional peak feature appearing in some of the spectra. This peak feature could not be correlated to any particular damage effect and may be influenced by several factors such as sample thickness and irradiation dose rate.

In order to compare the effect of fluorescence loss against radiation dose for the different scintillator types, a ratio between the integrated spectra over 350-500 nm, for the irradiated and un-irradiated regions were computed. A crude exponential fit was implemented to the data in order to provide a rough visual guide of the overall trend. This plot is shown in figure 3. The different scintillators perform within a 20% variation of each other. EJ208 exhibits the least fluorescence ratio loss, with UPS923A scintillators performing comparably against it, particularly at the higher dose exposures. The TileCal scintillators perform well against loss at the low doses, but lose light much faster at higher doses. After 25 MGy, EJ200 performs on par with EJ208 and UPS923A.

The results for EJ260 are not a true performance indicator as a very small component of the wavelength-shifted fluorescence was measured. In the Tile Calorimeter however, scintillation light is collected by wavelength shifting optical fibers which then lead to photo-multiplier tubes. The fibers presently used, Y11 fibers from Kuraray, have an absorption range of 420-450 nm and peak at 430nm. EJ260 therefore will not couple well to these fibers.

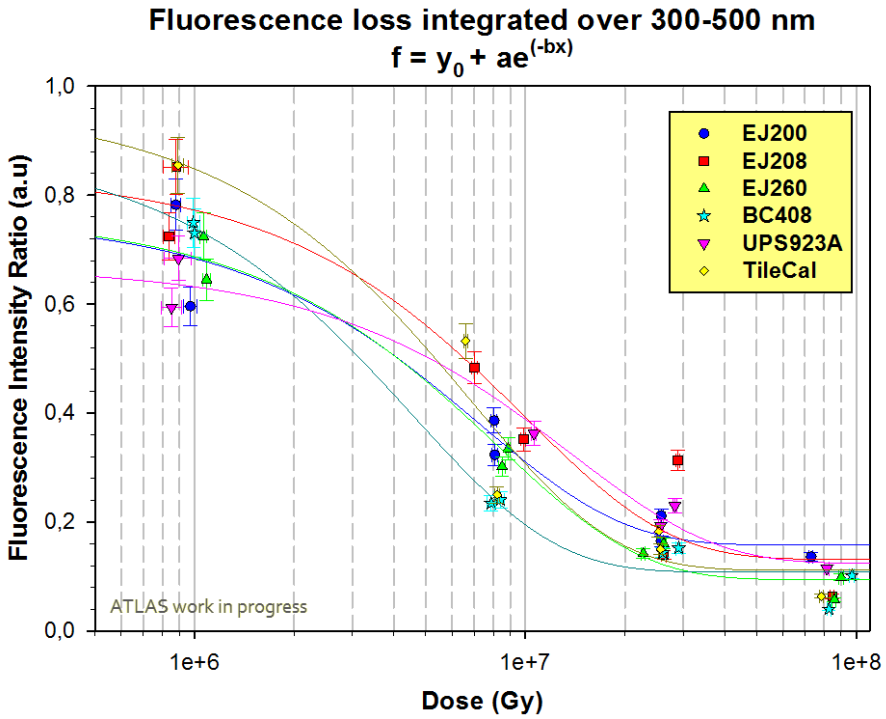

Figure 3: Ratio of fluorescence integrated over 300-500 nm for irradiated and un-irradiated regions of plastic scintillator samples.

5. Conclusion

The effect of 6 MeV proton induced radiation damage to the fluorescence capability of 350 μm thin PVT and PS based plastic scintillators was investigated. Fluorescence was measured off the surface of the samples using a 229 nm laser excitation. The following results were deduced:

- Fluorescence loss increases exponentially with increasing dose exposure.
- For doses below 1 MGy, a portion of the fluorescence arising from the added fluors is lost. Since a decrease in absorption corresponding to the fluors occurs for this dose as well, a bleaching-like effect may be the cause of damage to the fluors.
- As the dose exposure progresses to 8 MGy and above, fluorescence loss occurs for both the base and fluor emission regions. This ties in with Raman studies in [5], indicating that structural damage occurs to the polymer base. Colour centres or free radicals formed through the damage processes compete for absorption of base emitted light as well as some visible light over the lower blue region [5].
- At 70-90 MGy, minimal fluorescence over the fluor emission region is observed, whilst the base component of emission is lost. The different scintillator grades perform within a 20% variation of each other, with EJ208 exhibiting the least loss in fluorescence ratio.

A large component of the fluorescence light measured in this experiment was un-shifted light from scintillation off the polymer base since the geometry of the set-up minimised spatial interaction. Within the TileCal, scintillators employed are much larger and bulk effects play an important role in the scintillators performance. Whilst studying the thin samples give an indication of the finer damage mechanism at play, a study of the bulk effects is imperative for choosing the replacement candidate for the TileCal. Future work will therefore study the attenuation length and transmission of scintillation light in thick scintillators. Damage to the overall light loss in scintillator-fiber coupled systems will also be investigated. In addition, investigations using neutron irradiation are ongoing, and a new inorganic LYSO scintillation crystal is being added to the study.

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References

[1] ATLAS Collaboration 2008 *The ATLAS Experiment at the CERN Large Hadron Collider* (IOP Publishing and SISSA)

[2] Knoll G F 1999 *Radiation Detection and Measurement, Third Edition* (Michigan: John Wiley & Sons Inc.) chapter 8 pp 220-222

[3] Zorn C 1993 A pedestrian's guide to radiation damage in plastic scintillators *Radiat. Phys. Chem.* **41** 37-43

[4] Jivan H *et al* 2015 *Radiation hardness of plastic scintillators for the Tile Calorimeter of the ATLAS detector*, in Proceedings of SAIP2014, (University of Johannesburg), pp. 199 - 205. ISBN: 978-0-620-65391-6.

[5] Jivan H *et al* 2015 Radiation hardness of plastic scintillators for the Tile Calorimeter of the ATLAS detector *J. Phys.: Conf. Ser.* **623** 012016

[6] ELJEN Technology 2013 *Products: Plastic Scintilators* [Online]. Available: http://www.eljentechnology.com/index.php/products/plastic-scintillators.

[7] Saint-Gobain 2015 *Plastic Scintilators Specifications* [Online]. Available: http://www.crystals.saint-gobain.com/Plastic_Scintillators.aspx.

[8] ScintiTech 2014 *Products: Plastic Scintillators* [Online]. Available: http://www.scintitech.com/CompanyPage.aspx?MenuId=39&MainId=3.

[9] Karyukhin A *et al* 1996 *Injection molding scintillator for ATLAS Tile Calorimeter* ATL-TILECAL-96-086, ATL-L-PN-86

[10] Jivan H 2016 *Proton induced radiation damage studies on plastic scintilators for the Tile Calorimeter of the ATLAS detector* (Master's dissertation) [Online]. Available: http://wiredspace.wits.ac.za/handle/10539/21672