Radiation hardness of plastic scintillators for the Tile Calorimeter of the ATLAS detector

H Jivan1,2, E Sideras-Haddad1,2, R Erasmus1, S Liao1, M Madhuku1, G Peters1,2, K Sekonya1,3, O Solvyanov4

1: School of Physics, University of the Witwatersrand, Johannesburg 2050, South Africa
2: DST-NRF Centre for Excellence in Strong Materials
3: iThemba LABS, North, Empire Road, Braamfontein 2000, Johannesburg.
4: European Organization for Nuclear Research (CERN), CH-1211 Genève 23, Suisse

Email: harshna.jivan@gmail.com

Abstract The radiation damage in polyvinyl toluene based plastic scintillator EJ200 obtained from ELJEN technology was investigated. This forms part of a comparative study conducted to aid in the upgrade of the Tile Calorimeter of the ATLAS detector during which the Gap scintillators will be replaced. Samples subjected to 6 MeV proton irradiation using the tandem accelerator of iThemba LABS, were irradiated with doses of approximately 0.8 MGy, 8 MGy, 25 MGy and 80 MGy. The optical properties were investigated using transmission spectroscopy and light yield analysis whilst structural damage was assessed using Raman spectroscopy. Findings indicate that for the dose of 0.8 MGy, no structural damage occurs and light loss can be attributed to a breakdown in the light transfer between base and fluor dopants. For doses of 8 MGy to 80 MGy, structural damage leads to possible hydrogen loss in the benzene ring of the PVT base which forms free radicals. This results in an additional absorptive component causing increased transmission loss and light yield loss with increasing dose.

1. Introduction
The Tile Calorimeter of the ATLAS detector, is a hadronic calorimeter responsible for recording the energy and trajectory of hadrons, taus as well as jets of quarks and gluons that result from the proton-proton collisions within the Large Hadron Collider of CERN. Plastic scintillators form an integral component [1] of this calorimeter due to their ability to undergo fluorescence whereby incident radiation causes electronic excitations within the scintillator material which then relax back to their ground state through the emission of light [2]. Plastic scintillators in specific are chosen for their properties of high optical transmission and fast rise and decay time. This enables efficient data capture since fast signal pulses can be generated [2]. In addition, plastic scintillators are easier to manufacture as compared to inorganic crystals which require special growing methods, and are more cost effective [3] for covering the typically large detector areas. The main problem encountered by plastic scintillators however, is radiation damage incurred due to their interaction with the ionizing particles to be detected. This damage causes a significant decrease in the light yield of the scintillator and introduces an error into the time-of flight data acquired.

As part of the phase two upgrade of the Tile Calorimeter planned for 2018, a comparative study is underway which examines the radiation hardness of several plastic scintillators. The results will aid in
choosing the best grade for replacing the current crack scintillators within the Gap region of the Tile Calorimeter as well as provide insight into the type of damage that occurs. In these proceedings, we present preliminary results for EJ200, a polyvinyl toluene based plastic obtained from ELJEN Technologies.

2. Scintillator material under study
Plastic scintillator EJ200 is composed of a polyvinyl toluene base and 3% of added organic fluors. The absorption peak of the fluors coincide with the emission peak of the base. This allows for the fluors to absorb light emitted by the base and re-emit it at higher wavelengths, thus preventing re-absorption within the base. As a result, the scintillator becomes more transparent to its own light and the light output is boosted. Some important properties of EJ200 are listed in table 1 below.

| Table 1: Properties of plastic scintillator EJ200 as provided by the manufacturers [4] |
|-----------------------------------------------|
| EJ200                                         |
| Light Output (% Anthracene)                   | 64 |
| Wavelength of maximum emission (nm)           | 425 |
| Rise time (ns)                                | 0.9 |
| Decay time (ns)                               | 2.1 |
| Refractive index                              | 1.58 |

3. Experimental Procedure
The tandem accelerator of iThemba LABS in Gauteng was used to irradiate samples with 6 MeV protons. To ensure that the study simulated a similar type of particle-scintillator interaction as observed in the Tile Calorimeter, protons would be required to pass through the samples whilst imparting energy primarily through ionisation losses. In this manner, only electronic stopping is taking place while nuclear stopping which is responsible for dislocations is prevented. In order to ensure this, SRIM simulations were performed to determine the stopping range of 6 MeV protons within the plastic material. Based on these simulations, several samples were cut and polished to dimensions of 0.5 cm by 0.5 cm, with thickness of ~350 μm. A polishing procedure based on standard metallographic techniques was employed. Samples were irradiated to doses of 0.8 MGy, 8 MGy, 25 MGy and 80 MGy. A set of magnetic quadrupole triplets was used to focus the proton beam to a spot size of ~20-30 μm and the beam was then scanned in the x and y plane to achieve an irradiated area of approximately 1.8 mm by 1.8 mm.

The effects of proton damage on the optical properties of the samples were characterised by conducting light transmission and light yield studies. Transmission spectroscopy was conducted using the Varian Carry 500 spectrophotometer. Light transmission was measured relative to transmission in air over a range of 200-800 nm. Transmission spectra were collected prior to irradiation as well as over several days after. The scintillator light yield in response to 0.5 MeV gamma rays emitted by a $^{90}$Sr source was measured using the light box set-up at CERN. In these experiments, the sample was coupled to two Kuraray Y-11(200) optical fibers which were connected to a standard Tile Calorimeter photomultiplier tube. Light yield experiments were conducted several weeks after irradiation thus providing time for partial recovery in the scintillators.

The structural damage undergone by the scintillators were then investigated using Raman spectroscopy. Raman spectra were obtained for the un-irradiated control samples as well as on the 0.8 MGy and 8 MGy irradiated samples using the Horiba Jobin-Yvon Raman spectrograph. An Argon laser was used to provide a 515 nm excitation wavelength. Raman studies were conducted 3 days, 10 days and 4 weeks after irradiation.
4. Light Transmission Results and Analysis
The ratio of transmission in each irradiated EJ200 sample relative to its transmission before irradiation for several doses as a function of wavelength is shown in Fig. 1 below. Absorption in the PVT base occurs over 240 nm to 300 nm whilst absorption in the fluors is expected to occur over 270 nm to 400 nm. For the dose of 0.8 MGy, a transmission dip at lower wavelengths is observed, followed by a peak over 320–400 nm. This feature indicates that a competitive absorptive process has developed between the primary and secondary fluors.

As the dose increases, we observe the formation of an absorptive tint whereby the absorption drop-off shifts to higher wavelengths. This implies that additional absorbing species such as free radicals are forming within the scintillator and leads to a further decrease in light emitted by the scintillator. This additional absorption component may also reduce the attenuation length of the scintillation light as described by C. Zorn (1993) [5].

5. Light Yield Results and Analysis
Experiments conducted on the light output from the scintillator samples responding to 0.5 MeV gamma rays emitted from $^{90}$Sr were repeated thrice per sample and averaged. The light response in the irradiated samples as a ratio to light output in an un-irradiated control sample as a function of irradiation dose is shown in Figure 2. There appears to be a linear decreasing trend in light loss with increasing orders of magnitude in dose. However, these results are very preliminary as geometric considerations in the experimental set-up need to be accounted for. This may account for the large variation in light loss seen between two samples irradiated to a similar dose of ~ 0.9-1 MGy.
Figure 2: Relative light yield in EJ200 samples as a function of irradiation dose.

6. Raman Spectroscopy Results and Analysis

Raman spectra were obtained for the 0.8 MGy samples, 8 MGy samples as well as for un-irradiated control samples. The samples irradiated to 0.8 MGy maintained their structure and very little damage was observed. In the 8 MGy sample, an increased amount of background fluorescence could be observed which may be due to free radicals interacting with the excitation light. Raman spectroscopy could not be performed for the high dose of 80 MGy because this background fluorescence becomes too large to obtain an experimentally viable spectrum. Figure 3 shows the background subtracted Raman spectrum for the 8 MGy irradiated sample taken over several days.

Figure 3: Background subtracted Raman spectra for 8 MGy irradiated EJ200. N.B Spectra overlap but intensities have been shifted in order to have better visible representation.
The peaks observed are allocated to the characteristic functional groups or vibrational groups using the Raman Peak assignment datasheet from Horiba Jobin-Yvon. The peak assignment is summarized in Table 2. An additional peak (X) was found to form after irradiation but disappeared over time as the scintillator healed. In order to gauge changes in the amount of specific species present, the intensities of the various peaks were plotted relative to peak 13 which represents the C-C bond typically found in benzene rings. These results are shown in Figure 4.

Table 2: Allocation of peaks to characteristic functional groups or vibrational groups.

| Functional Group/ Vibration                          | Peak Assignment |
|------------------------------------------------------|-----------------|
| δ(C-C) aliphatic                                     | 20-21           |
| ν(C-C) alicyclic or aliphatic chain vibrations       | 9-12, 14-19, X  |
| ν(C-C) aromatic ring chain vibrations               | 13              |
| δ(CH3)                                              | 8               |
| δ(CH2) or δ(CH3) asymmetric                         | 7               |
| ν(C=C)                                               | 6               |
| ν(C-H)                                               | 3-4             |
| ν(=(C-H))                                            | 1-2             |

The ratio of species typically found in the vinyl backbone of the sample versus that in the benzene ring shows an increase after irradiation. A small amount of additional alicyclic rings and aliphatic chains are found with the additional peak X being typical of these bonds. These bonds represent single carbon rings and chains stripped of hydrogen. This indicates that structural damage occurs within the scintillator whereby bonds in the benzene ring are broken. Hydrogen may be lost by the benzene rings to form free radicals. After 4 weeks of healing, the scintillator appears to substantially recover its structure.

![Figure 4](image.png)

Figure 4: Plot of the intensities of peaks relative to peak 13 representing the C-C aromatic ring bond.
7. Conclusion
According to the results obtained in this study, irradiation to high doses in plastic scintillator EJ200 using 6 MeV protons causes radiation damage and leads to a reduced performance in its scintillation capabilities. At a dose of approximately 0.8 MGy, the structure of the scintillator remains robust, however light loss is still observed. This can be accounted for by noting that light transmission indicates a competitive absorptive process developing between the primary and secondary fluors.

At a dose of 8 MGy, more light loss is observed and the scintillator becomes less transparent to its own light. The formation of an absorptive tint indicates that free radicals may be absorbing light that would otherwise be collected from the scintillator. These free radicals may arise from hydrogen that is lost through bonds breaking within the benzene rings of the polyvinyl tolouene base and hence leading to additional aliphatic chains and alicyclic rings observed in the Raman spectra. The Raman spectra further indicate that after 4 weeks, the scintillator significantly recovers its structure. As the irradiation dose is increased, a greater loss to light transparency is observed. This can also be visibly noticed in the increased yellowing of the irradiation spot among samples with progressing higher doses. Increasing dose leads to greater structural damage and leads to a reduction in the light output of the scintillator.

Overall, EJ200 can be considered as substantially radiation hard since for a dose of 0.8 MGy, it still maintains its structural composition, remains fairly transparent and its light output is still greater than 50% of its original capability. It is only at doses higher than 8 MGy that permanent structural damage occurs. The highest dose currently expected within the Tile Calorimeter over the next several years is 10 kGy and therefore EJ200 may be a potential candidate for consideration during the 2018 replacement of the GAP scintillators.

8. Upcoming work
This study is being carried out on several other plastic scintillators including PVT based EJ208, EJ260 and BC-408 obtained from ELJEN technologies and Saint Gobain Crystals respectively. These commercial scintillators will be compared to the polystyrene based scintillators presently used within the Tile Calorimeter. In addition to the techniques looked at in this paper, healing in the scintillators as well as the effect of dose rate on the damage induced are being looked at. Electron Paramagnetic resonance (EPR) and nuclear magnetic resonance (NMR) studies are also being conducted in order identify the types of free radicals formed due to radiation damage.

9. 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] Chen M 2011 Queen's University, PHYS 352: Measurement, Instrumentation and Experiment Design [Online]. Available: http://www.physics.queensu.ca/~phys352/lect19.pdf.
[4] ELJEN Technology 2013 Products: Plastic Scintillators [Online]. Available: http://www.eljentechnology.com/index.php/products/plastic-scintillators.
[5] Zorn C 1993 A pedestrian’s guide to radiation damage in plastic scintillators Radiat. Phys. Chem. 41 37-43