A comparative study of the radiation hardness of plastic scintillators for the upgrade of the Tile Calorimeter of the ATLAS detector

S. Liao, R. Erasmus, H. Jivan, C. Pelwan, G. Peters and E. Sideras-Haddad
School of Physics, University of the Witwatersrand, Johannesburg 2050, South Africa
E-mail: shell-may.liao@cern.ch

Abstract. The influence of radiation on the light transmittance of plastic scintillators was studied experimentally. The high optical transmittance property of plastic scintillators makes them essential in the effective functioning of the Tile calorimeter of the ATLAS detector at CERN. This significant role played by the scintillators makes this research imperative in the movement towards the upgrade of the tile calorimeter. The radiation damage of polyvinyl toluene (PVT) based plastic scintillators was studied, namely, EJ-200, EJ-208 and EJ-260, all manufactured and provided to us by ELJEN technology. In addition, in order to compare to scintillator brands actually in use at the ATLAS detector currently, two polystyrene (PS) based scintillators and an additional PVT based scintillator were also scrutinized in this study, namely, Dubna, Protvino and Bicron, respectively. All the samples were irradiated using a 6 MeV proton beam at different doses at iThemba LABS Gauteng. The radiation process was planned and mimicked by doing simulations using a SRIM program. In addition, transmission spectra for the irradiated and unirradiated samples of each grade were obtained, observed and analyzed.

1. Introduction
Plastic scintillators are materials that exhibit the property of luminescence when excited by ionizing radiation. This, together with their high optical transmission property makes them suitable for use within large particle detectors such as the ATLAS detector of the Large Hadron Collider (LHC) at CERN [3]. The plastic scintillators under scrutiny are found within the Tile calorimeter of the ATLAS detector. The Tile calorimeter is situated within the hadronic calorimeter of the ATLAS detector, which usually detects hadrons, quarks and gluons. It is 8 metres in diameter and 12 metres in length and makes use of steel as the absorber material, as well as the scintillation plates which are of interest in this research [4]. The steel plates act as an absorption medium and convert the incoming jets into a shower of particles. These showers tend to interact with the plastic tiles and the tiles absorb the energy from these particles and emit the energy in the form of light. The light that is produced within the scintillators is then guided by wavelength shifting fibres and read out through the photomultiplier tubes (PMTs) [4]. The performance of the Tile calorimeter therefore has a direct impact on signatures involving hadrons, jets and missing transverse energy. However, the main problem encountered by these scintillators is radiation damage due to the highly ionizing nature of the particles that are being
detected. Thus, due to the anticipated increase in beam energy, there is bound to be an increase in the amount of radiation that the tiles are exposed to. As a result, the need for the use of the most radiation hard plastic scintillators is mandatory in the movement towards upgrading the Tile calorimeter of the ATLAS detector.

2. The scintillation mechanism

The plastic scintillators used in this study were organic Polyvinyl Toluene (PVT) based scintillators doped with fluors. The fast response and low cost production of organic scintillators such as the PVT makes them a rational choice for use in large detectors. They are made up of long chain vinyl toluene molecules. Toluene consists of a benzene ring bonded to a methyl (CH3) and a vinyl group (CH2-CH-) [6]. The benzene ring is a common feature in plastic scintillators and it is characterized by delocalization of 3 π-bonds [1]. The chemical bonds present in a benzene ring include σ-bonds which are in the plane and π-bonds which are out of the plane and overlap. The π-bonds give rise to a cloud of electrons above and below the molecular plane and these π electrons are completely delocalized. Figure 1 below shows the overlap of the π-bonds which give rise to the delocalized electrons.

![Figure 1. Schematic diagram of the π-bond overlap](image)

![Figure 2. Electronic energy levels of an organic molecule](image)

These delocalized electrons are prone to excitation by radiation, which is essential in the scintillation process. In fact, it is the de-excitation of these delocalized π electrons which result in luminescence. Figure 2 above shows the π-electronic energy levels of an organic molecule. When particles strike the surface of such a scintillator, the radiation is absorbed and results in a molecular excitation to the first excited singlet state. This is almost instantly followed by de-excitation back to the ground state, which is called fluorescence (scintillation). In addition,
plastic scintillators are doped with organic fluors: Primary fluor and Secondary fluor. These organic fluors are compounds which absorb the base scintillation and re-emit it at longer wavelengths.

3. Methods

3.1. Modelling the radiation environment

Within the Tile calorimeter, incoming jets carry high energy and interact with the plastic scintillators by imparting energy through this ionization process, and such interactions are essential in understanding the damage mechanism within the target. This calls for a method that one can use to model the radiation environment prior to the actual radiation process in order for effective research results. This modelling was done using SRIM simulations. When the ions interact with the target atoms, they do not get stopped within the material. That is, the ions propagate straight through the material without stopping, hence the need to ensure that the ion beam that will be used during the radiation process also pass straight through the scintillators without stopping. SRIM simulations were then conducted in order to find out whether the proton beam will pass straight through the 350 um thick Polyvinyl Toluene based plastic scintillators, as this was the thickness that we anticipated to cut the samples into. The necessary information that is extracted from the SRIM output is that the 6 MeV proton beam stops at a distance of 474.17 um. Since the samples which are used in this research are 350 um thick, this data tells us that the 6 MeV proton beam will pass straight through the plastic scintillators without stopping. Furthermore, TRIM establishes collisions between the ions and target atoms and thus give information about the energy lost by the protons during ionization.

From this knowledge, we were able to plan the beam time and beam currents needed to achieve the anticipated doses using the following relation:

$$ R = \frac{it \times E(lost)}{q \times m} \quad (1) $$

Thus from the simulations, the average energy of the transmitted protons through a 350 um thick PVT target is approximately 2.8MeV, which implies that the energy lost by the 6MeV protons is given by:

$$ 6MeV - 2.8MeV = 3.2MeV \quad (2) $$

3.2. Sample preparation

Sample preparation was carried out within the physics building Materials Preparation Lab at the University of the Witwatersrand. Initially, the plastic scintillator samples were of dimensions of approximately 200 mm X 25 mm X 10 mm in length, width and thickness, respectively. For the irradiation process the samples needed to be sectioned and cut to dimensions 5 mm X 5 mm X 0.35 mm in length, width and thickness, respectively. Following the cutting, the samples were mounted onto aluminum holders and polished down to the required thickness using a Struers Metallographic polishing machine. After the samples were polished, their thicknesses were measured and documented and it was found that there was a variation in the thicknesses of the samples. That is, they ranged between 300 um and 380 um. Simulations were then done again for this range of thicknesses but in 10 um intervals in order to ensure accurate beam time planning.
4. Experimental procedure

4.1. Radiation process
The radiation of the PVT based plastic scintillators was done using the Tandem accelerator found at iThemba LABS Gauteng. A 6 MeV proton beam was used during the irradiation process. Each brand of PVT based plastic scintillator was exposed to four different doses, a high dose (80 MGy), an intermediate dose (25 MGy), a medium dose (8 MGy) and a low dose (0.8 MGy).

4.2. Transmission spectroscopy
Immediately after irradiation the samples were subject to transmission spectroscopy. That is, data for light transmittance of the PVT based plastic scintillators was taken. This was done using The Varian Cary 500 spectrophotometer which is available within the physics building of the University of the Witwatersrand. Data for transmission data was taken and plotted as percentage transmission versus the wavelength of light. The transmission data obtained was plotted in order to observe and analyze the effects of radiation damage on the light transmittance of the plastic scintillators.

5. Results and analysis
The data was input and plotted using ROOT plot as it is the prominent plotting program used at CERN. In order to gauge whether the decrease in % light transmission increased with the radiation dose, graphs of % transmission versus wavelength for each grade at different doses were plotted, as seen in the case of EJ 208 (Figure 3). Also, in order to observe whether the samples showed some recovery or not following the radiation process, graphs of % transmission versus wavelength for each grade for transmission taken on different days were also plotted, as seen in the case of EJ 208 in Figure 4.

*Figure 3. A plot of Transmission versus wavelength for EJ 208 for all doses*
Figure 4. A plot of transmission versus wavelength for EJ 208 on different days

| Sample | Dose (MGy) | % Trans. loss | Sample | Dose (MGy) | % Trans. loss |
|--------|------------|---------------|--------|------------|---------------|
| EJ 200 | 80         | 42.9          | Protvino | 80         | 60.8          |
|        | 25         | 28.6          |         | 25         | 34.8          |
|        | 8          | 14            |         | 8          | 7.4           |
|        | 0.8        | 3.9           |         | 0.8        | 3.3           |
| EJ 208 | 80         | 29.1          | Dubna   | 80         | 51.3          |
|        | 25         | 14.9          |         | 25         | 35.1          |
|        | 8          | 4.7           |         | 8          | 26.6          |
|        | 0.8        | 2.5           |         | 0.8        | 5.5           |
| EJ 260 | 80         | 44.8          | Bicron  | 80         | 45.5          |
|        | 25         | 15.5          |         | 25         | 39.5          |
|        | 8          | 14.3          |         | 8          | 11.5          |
|        | 0.8        | 6.6           |         | 0.8        | 8.7           |

Table 1. Table showing the % transmission loss for all samples at a wavelength of 420 nm, over the different doses
In Figure 3 as each sample is exposed to radiation, the absorptive tint evident in the unirradiated graph tends to shift towards longer wavelengths as the dose is increased. In addition, a decline in percentage transmission is evident as well and the percentage transmission loss increases with increase in dose. Also, Figure 4 shows that indeed the samples do undergo some form of recovery after the radiation process. Between the day of irradiation and 2 or 3 days after irradiation, there is a substantial amount of increase in percentage transmission, indicating the sample recovery. The following weeks also indicate some healing, but small in comparison to the first few days following the irradiation process. The recovery was also evident in the visual changes of the sample irradiation spots. For very high doses, the dark yellow radiation spot faded to a normal yellow and for the lower doses the spot was nearly invisible even on the days of irradiation.

Table 1 shows the percentage loss in transmission for each sample over each dose in comparison to its unirradiated. That is, for each dose at a wavelength of 420 nm, the percentage transmission loss was calculated as follows:

\[
\% \text{ transmission loss} = \% \text{ transmission of unirradiated} - \% \text{ transmission of irradiated} \quad (3)
\]

From the table, one can see that the PS based plastic scintillators (protvino and dubna) undergo the most decrease in transmission for most of the doses. Also, the bicron shows high percentage loss in transmission for all the doses. On the other hand, the PVT based scintillators provided by ELJEN technologies show the least amount of decline in percentage transmission for all the doses in comparison to all the other grades. On the other hand, Table 2 shows the difference in percentage transmission between two different days that transmission data was taken. This was done for all the EJ 200, EJ 208 and EJ 260 for only two specific doses. For the higher dose EJ 208 showed the most amount of healing as a large difference in transmission is seen. For the smaller dose, EJ 200 reflected the most amount of healing in comparison to the others.

6. Conclusions

Even though plastic scintillators generally possess overall commendable properties, their vulnerability when exposed to high doses of radiation is a major predicament. Unfortunately, all types of scintillators are affected by radiation. Studies done by Bross [2] indicate that light yield losses in plastic scintillators are not due to the dopant degradation, but rather to the damage within the base matrix. In addition, according to Torrisi [7], when PVT based polymers are exposed to high radiation doses, their chemical bonds are broken and their polymer properties are changed. Apparently, at the breaking points of the carbon and hydrogen bonds, free radicals are formed and hydrogen degassing occurs, resulting in a material which is rich in carbon. The formation
of free radicals is bound to affect the scintillating mechanism of the plastic scintillators as they tend to absorb fluorescent light thus limiting the light yield of the scintillators.

In our transmission results, we found that indeed an increase in radiation results in a decline in light transmission for all of the different grades. This could be because the dopants are no longer emitting the base scintillation but are rather absorbing it instead. When comparing the transmission spectra of each dose with its unirradiated in the wavelength range that is read out by the optical fibres of the ATLAS detector, we found that the PVT based scintillators provided by ELJEN technologies showed the least amount of decline in percentage transmission in comparison to the other scintillators. This is evident in the data in Table 1. Amongst the PVT based scintillators, we found that EJ 208 showed the least amount of decline in percentage transmission for most of the doses. On the other hand, EJ 260 showed the highest amount of decline in percentage transmission in comparison to the other two brands.

Also, the annealing of a sample after the process of irradiation plays an important role in establishing whether a material is radiation hard or not. From the transmission data collected on different days in order to observe annealing, we found that the plastic scintillators do indeed undergo some healing after radiation. From comparing the healing of two different doses we found that EJ 208 and EJ 200 both experienced a substantial amount of healing in comparison to EJ 260. Also, it was also noted that for all the grades, the most amount of healing occurred within the first few days after irradiation in comparison to weeks later.

Moreover, from these results and observations, it is clear that even though EJ 200 showed the best transmission properties at the start, after irradiation, EJ 208 reflected more radiation hardness. This is supported by the fact that it showed the least amount of decrease in percentage light transmission as well as also healed a substantial amount in a short period of time. Light yield results are currently being done and will further aid in the recommendation of the best plastic scintillator grade.

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