Neutron induced radiation damage of plastic scintillators for the upgrade of the Tile Calorimeter of the ATLAS detector.

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Abstract. With the prediction that the plastic scintillators in the gap region of the Tile Calorimeter will sustain a significantly large amount of radiation damage during the HL-LHC run time, the current plastic scintillators will need to be replaced during the phase 2 upgrade in 2018. The scintillators in the gap region were exposed to a radiation environment of up to 10 kGy/year during the first run of data taking and with the luminosity being increased by a factor of 10, the radiation environment will be extremely harsh. We report on the radiation damage to the optical properties of plastic scintillators following irradiation using a neutron beam of the IBR-2 pulsed reactor in Joint Institute for Nuclear Research (JINR), Dubna. A comparison is drawn between polyvinyl toluene based commercial scintillators EJ200, EJ208 and EJ260 as well as polystyrene based scintillator from Kharkov. The samples were subjected to irradiation with high energy neutrons and a flux density range of $1 \times 10^6$–$7 \times 10^6$. Light transmission, Raman spectroscopy, fluorescence spectroscopy and light yield testing was performed to characterize the damage induced in the samples. Preliminary results from the tests done indicate a minute change in the optical properties of the scintillators with further studies underway to gain a better understanding of the interaction between neutrons with plastic scintillators.

1. Introduction
The ATLAS detector is at one of the four collision points for protons on the Large Hadron Collider (LHC) ring together with CMS, ALICE and LHCb detectors. It is used to search for new particles through high energy proton-proton collisions at the LHC of CERN and also perform unprecedented precision measurements. The detector consists of six different detecting subsystems that are arranged in layers around the collision point to record the path, momentum and energy of the particles in order for the particles to be individually identified[1]. Amongst the detecting subsystem, there is the calorimeter subsystem that consists of the tile calorimeter. The tile calorimeter is a hadronic calorimeter responsible for detecting hadrons. It consists of a central barrel and 2 extended barrels with each barrel containing 64 modules that are made of a matrix of steel plates and plastic scintillator tiles sandwiched between them. The steel plates act as an absorber medium that converts the incoming hadrons into a “shower” of particles. The
plastic scintillator tiles then absorb the energy of the particles and fluorescence to emit light. The light from the scintillators is passed through wavelength shifting (WLS) optical fibres and is detected by photomultiplier tubes. The signal is further processed using readout electronics in order to digitize the data for further analysis [2] [3].

In addition to the plastic scintillators found within the modules of the calorimeter, additional scintillators are found in the gap region. The gap region is the area between the central and extended barrels. During the first run of data taking, the scintillators in the gap region were exposed to a radiation environment of up to 10 kGy/year. It is predicted that during the high luminosity (HL)-LHC run time, the scintillators in the Gap region will sustain a significantly large amount of radiation damage and will require replacement. This prediction has led to the comparative study of proton induced radiation damage on plastic scintillators conducted by H Jivan [2] [4], C Pelwan [5] and S Liao [6].

To further extend the project on the comparative study, neutron irradiation and damage assessment studies on the same plastic scintillator samples is being conducted. Neutron interaction with matter makes this study interesting since unlike with proton irradiation where the interaction with the plastics was through direct ionization, with neutron irradiation the interaction will be through indirect ionization. Neutron bombardment on materials creates a collision cascade within the material that results to point defects and dislocations. The collisions cause a massive transfer of kinetic energy to the lattice atom that has been displaced from its lattice site, becoming what is known as the primary knock-on atom (PKA). The knock-on atoms then lose energy with each collision and that energy therefore ionizes the material [7]. The study follows the same experimental procedures as that of the proton induced radiation damage study with a few minor changes to better suit the study.

2. Scintillation mechanism

Four plastic scintillator grades were under study, three of which were obtained from ELJEN Technologies and one from ISMA, Kharkov. The plastic scintillator grades under study are the EJ200, EJ208, EJ260 and the UPS-932A type. The three plastic scintillators obtained from ELJEN technologies are composed of a polyvinyl toluene base and 3% added organic fluors [1], the fourth plastic scintillator grade is the UPS-932A type obtained from ISMA, Kharkov and is composed of polysterene base. Polyvinyl toluene consists of long chains of vinyl toluene molecules that encompass a benzene ring ($C_6H_4$) bonded to a methyl group ($CH_3$) and a vinyl group ($CH_2 − CH −$). Information on the exact composition of the doped fluors are not made public by the manufacturers. Table 1 below has some important properties of the Eljen samples listed.

| Table 1. Properties of Eljen scintillators [8]. |
|-----------------------------------------------|
| Light Output (% Anthracene) | EJ200 | EJ208 | EJ260 |
| Wavelength of maximum emission (nm) | 425 | 435 | 490 |
| Rise time (ns) | 0.9 | 1.0 | - |
| Decay time (ns) | 2.1 | 3.3 | 9.2 |
| Refractive index | 1.58 | 1.58 | 1.58 |

Organic scintillators have a basic scintillation mechanism that involves Foster energy transfer and self-absorption. They consists of one or two dopants [9]. The benzene ring within the scintillators determines the scintillation mechanism. The chemical bonds found within a benzene ring are: $\sigma$-bonds that are in the plane with bond angle 120° and are from $sp^3$ hybridization. The
other chemical bonds found are $\pi$-orbitals which are out of plane and overlap. The $\pi$-electrons are completely delocalized.

The molecule undergoes vibrational relaxation to the $S_{10}$ state after the scintillator has absorbed a photon or excitation by ionization. The $S_{10}$ excited state radiatively decays to the vibrational sub-levels of the ground state. The lifetime of the $S_{10}$ state is in the nanoseconds time range. The short lifetime allows for the fluorescence emission spectrum to be roughly a “mirror image” of the absorption spectrum. The Stokes shift is observed when the emitted photons have less energy than the $S_{00} - S_{10}$ phase transition. There is no $S_2 - S_0$ emission, thus there is an internal non-radiatively de-excitation occurring within the scintillator taking place in the picoseconds time range. The excited triplet state cannot decay to the ground state as a result angular momentum selection rules, it therefore results in a delayed fluorescence and phosphorescence [9].

![Figure 1. Energy level diagram of an organic molecule with $\pi$-electron structure, adapted from[9].The singlet states with spin 0 are labelled $S_{ab}$ and the triplet states of spin 1 are labelled $T_{ab}$ where $a$ indicates the excitation level and $b$ indicates the vibrational state of that level.](image)

3. Experimental Procedure

Several samples of each plastic scintillator grade were cut and polished to dimensions 20 mm by 20 mm by 6 mm thickness at the Dzhelepov Laboratory of Nuclear Problems (DLNP) at JINR. Special sample holders were made to accommodate the samples for irradiation. Channel 3 of the IBR-2 pulsed reactor at the Frank Laboratory of Neutron Physics (FLNP) at JINR was used to irradiate the samples [10]. The samples were subjected to irradiation with a beam of fast neutrons with a wide energy range of up to 10 $MeV$ for 337 hours. The reactor was operating at an average power of 1875 $kW$. The samples were placed at three different positions from the reactor core to expose them to different neutron fluences in order to achieve various doses. The neutron flux density ranged between $1 \times 10^6$ – $7.7 \times 10^6 n/cm^2/s$.

The Monte Carlo N-Particle (MCNP) 5 [11] code was used to simulate neutron transport through plastic scintillators and to determine the dose rate. The Monte Carlo method is used to simulate statistical processes theoretically, in particular complex problems that cannot be solved/modelled using computer codes that use deterministic methods. The MCNP code is used to simulate neutron, photon and electron or coupled neutron/photon/electron transport. The code is in 3-D and is capable of tracking up to 34 particles and 4 light ions. The code takes into account the absorption and the moderation of the neutrons as they travel though matter. Table 2 below shows the neutron flux density and neutron fluences at the various positions.
Table 2. Neutron flux density and neutron fluences at the various positions.

| Sample position | Flux density \((n/cm^2/s)\) | Fluence \((n/cm^2)\) |
|-----------------|-----------------------------|---------------------|
| 1               | \(1 \times 10^6\)           | \(1.2 \times 10^{12}\) |
| 2               | \(3.6 \times 10^6\)         | \(3.6 \times 10^{12}\) |
| 3               | \(7.7 \times 10^6\)         | \(9.4 \times 10^{12}\) |

Light spectroscopy was conducted using the Varian Carry 500 spectrophotometer to characterize the optical properties of the irradiated samples due to the damage of the neutron irradiation. The light transmission of the samples was measured relative to the transmission in air over a laser wavelength range between 200-800 nm.

The Raman spectroscopy techniques was used to investigate any structural damage undergone by the scintillators after irradiation. The Horiba Jobin-Yvon Raman spectrograph consists of an Argon laser used to provide a 515 nm excitation wavelength. Raman spectra were obtained for the un-irradiated control samples as well as on the irradiated samples to gauge the changes in the samples.

Fluorescence testing was measured with the aid of LabrAM HR Raman spectroscopy setup. A laser of wavelength of 229 nm with a power ranging from 3 \(\sim\) 5 mW was employed to provide enough energy for molecular excitation, which results in prompt light emission through luminescence. The laser is guided through a series of mirrors and optics in the setup, and is incident on the sample through the microscope aperture. As the sample fluoresces, the light emitted in the direction back up the aperture is collected by a detector and a differential wavelength spectrum is obtained.

Light yield testing was conducted at CERN. The samples were excited with a Sr-90 source that undergoes beta decay. As the sample fluorescence to emit light, the light is passed through four WLS optical fibres and detected by a PMT. The signal from the PMT is further processed through electronics and is digitized in the computer. For better contact between the sample and the fibres, profiles were placed along the sides of the sample to ensure the stability of the fibres.

4. Results and Discussion

Below are the spectra of the EJ200 samples. All the other scintillator grades behaved in the same manner as the EJ200 and therefore their results will not be discussed in detail.

Figure 2 shows the light transmission spectra of the EJ200. It is observed that at a wavelength of 400 nm the absorptive edge falls away completely and that the overall transmission of the grade remains unchanged even after irradiation. The transmission loss is observed at wavelength 450 nm as this corresponds to the peak absorption wavelength of the wavelength shifting optical fibres coupled with these scintillators within the Tile Calorimeter. Even at this wavelength, there is still no change observed in the samples with increasing doses.
Figure 2. Left: Light transmission spectra for the EJ200 samples. Right: Raman spectra for the EJ200 samples.

Raman spectra were obtained for the various irradiated samples as well as for un-irradiated control samples. The irradiated samples maintained their structure with no additional peaks being formed and no damage was observed. Figure 2 also shows the Raman spectra of the EJ200 samples for the irradiated and un-irradiated samples.

Figure 3 shows the fluorescence spectra of the EJ200 samples. Several fluorescence peak features are observed in the spectra. Peaks in the wavelength region of 300-375 nm correlate to fluorescence of the PVT/PS base. The "two-peak" feature is observed since the fluorescence is predominantly from the benzene ring structure. In the wavelength region of 375-500 nm, the fluorescence correlates to that of the fluor dopants. No change in fluorescence is observed in the spectra for the irradiated and un-irradiated samples.

Figure 3. Left: Fluorescence spectra for the EJ200 samples. Right: Light yield output signal versus neutron fluence.

The light yield of the scintillators is also shown in figure 3. The light yield of the samples was measured by testing their response to a Sr-90 source that undergoes beta decay. As the sample fluorescence to emit light, the light is passed through four WLS optical fibres and detected by
a PMT. The signal from the PMT is further processed through electronics and is digitized in the computer. This setup was aimed at mimicking what happens within the detector when the scintillators are measuring the energy of the particles. From the graph below, we observe no change for all the scintillator grades with irradiation.

5. Further work
During the autumn 2016 run of the IBR-2 reactor, composite scintillators were irradiated at higher neutron fluences than previously done. Composite scintillators were irradiated at higher neutron fluences range of $3.8 \times 10^{-12} - 1.8 \times 10^{14} \text{ n/cm}^2$. Raman Spectroscopy, Light yield measurements and Light transmission testing were performed on the irradiated samples. Light yield measurements were done at the Dzhelepov Laboratory for Nuclear Problems at the Joint Institute for Nuclear Research.

The light yield decreased by $\sim 28\%$ after the neutron irradiation of $1.8 \times 10^{14} \text{ n/cm}^2$. A decrease in the light transmission is also observed from the light transmission spectrum. In figure 4 on the left, we observe the change in the light transmission spectrum on the inset clearly. On the right of figure 4 we observe the light yield graph.

![Figure 4](image)

**Figure 4.** Left: Light transmission of the composite scintillator. Right: Light yield output of the composite scintillator.

No evident structural changes were observed after the irradiation from the Raman spectroscopy measurements. No additional peaks were formed after irradiation. Figure 5 shows the Raman spectra of the composite scintillator, the un-irradiated spectrum with the irradiated spectra above it.

![Figure 5](image)

**Figure 5.** Raman spectrum of the composite scintillators.
6. Conclusion
The radiation damage the plastics were exposed to is very low relative to that within the ATLAS detector. Structural and optical measurements performed to study the damage of the plastics for the first set of samples that were irradiated during the April 2016 run yielded no change. The study was further extended to higher fluences during the autumn 2016 run of the reactor. Changes in the transmission of the scintillators was observed at fluences $1.8 \times 10^{14} \text{n/cm}^2$. This is a good indication of what range of neutron flux the plastics need to be subjected to in order to start observing changes. Further analysis on the plastics is done and plans to work at even higher fluences are underway.

7. Acknowledgments
We would like to acknowledge the Frank Laboratory of Neutron Physics (FLNP) for making the IBR-2 reactor available for experiments and the staff involved. We also acknowledge SA-CERN consortium, NRF and SA-JINR for the financial support.

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