Neutron irradiation and damage assessment of plastic scintillators of the Tile Calorimeter

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Abstract. Following the comparative study of proton induced radiation damage on various plastic scintillator samples from the ATLAS-CERN detector, a study on neutron irradiation and damage assessment on the same type of samples will be conducted. The samples will be irradiated with different dose rates of neutrons produced in favourable nuclear reactions using a radiofrequency linear particle accelerator as well as from the SAFARI nuclear reactor at NECSA. The MCNP 5 code will be utilized in simulating the neutron transport for determining the dose rate. Light transmission and light yield tests will be performed in order to assess the radiation damage on the scintillators. In addition, Raman spectroscopy and Electron Paramagnetic Resonance (EPR) analysis will be used to characterize the samples after irradiation. The project aims to extent these studies to include radiation assessment damage of any component that processes the scintillating light and deteriorates the quantum efficiency of the Tilecal detector, namely, photomultiplier tubes, wavelength shifting optical fibres and the readout electronics. They will also be exposed to neutron irradiation and the damage assessed in the same manner.

1. Introduction
The ATLAS detector is used in particle physics experiments that are involved in the search for new particles through high energy proton-proton collisions at the Large Hadron Collider (LHC) of CERN. The tile calorimeter is part of the ATLAS detector, it is the hadronic calorimeter responsible for detecting hadrons, taus, and jets of quarks and gluons. The tile calorimeter consists of a central barrel and 2 extended barrels. Each barrel contains 64 modules that consists of a matrix of steel plates and plastic scintillators. The steel plates act as an absorber medium that converts the incoming jets 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 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\cite{1} \cite{2}.

Between the central barrel and extended barrels there is what is referred to as the Gap region. This region contains additional plastic scintillators that are radially distributed within the region. During the first run of data taking, the scintillators in the gap region were exposed to a radiation environment of up to 10kGy/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 during the 2018 upgrade. This prediction has led to the comparative study of proton induced radiation damage on plastic scintillators conducted by H Jivan\cite{1} \cite{3}, C Pelwan\cite{4} and S Liao\cite{5}.
To further extend the project on the comparative study, neutron irradiation and damage assessment studies on the same plastic scintillator samples will be conducted. The study will follow the same experimental procedures as that of the proton induced radiation damage study.

2. Scintillation mechanism

The plastic scintillator samples that will be studied are organic scintillators, these scintillators have a basic scintillation mechanism that involves Foster energy transfer and self-absorption. They consist of one or two dopants [6]. The scintillation mechanism of organic scintillators is determined by the physics and chemistry of the benzene ring. An organic scintillator scintillates despite its crystal form, whether it be in a liquid, a gas or imbedded in a polymer. 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.

Looking at the scintillation mechanism after the scintillator has absorbed the photon or excitation by ionization, the molecule will undergo vibrational relaxation to the \( S_{10} \) state. 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, in other words, they have the same spacing. The emitted photons have less energy than the \( S_{00} \) – \( S_{10} \) phase transition and that’s where the important Stokes shift is observed. 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 [7].

![Image of fluorescence absorption and emission spectra and scintillation mechanism](image.png)

Figure 1. A fluorescence absorption and emission spectra (left) and scintillation mechanism for an organic scintillator (right) [7].

3. Neutron physics

3.1. Neutron production

Neutrons can interact with both nuclei and electrons in matter, with the scattering cross-section of similar sizes for both. Since they can penetrate deeply into matter, they provide an ideal probe of bulk properties of matter which is ideal for our plastic scintillator study [8]. There are 2 fundamental mechanisms that can provide neutrons for slow-neutron scattering purposes in present day research facilities, namely, fission and spallation.
In nuclear physics, nuclear fission can either be a radioactive decay process or a nuclear reaction where a nucleus of an atom will split into smaller parts of lighter nuclei. In an induced nuclear fission reaction, a slow/thermal neutron is absorbed by a nucleus; Uranium-235 for example. The nucleus is turned into an excited Uranium-236 where the excitation energy is provided by the kinetic energy of the neutron plus the forces that bind the neutron. The Uranium-236 then splits into fast moving lighter nuclei/elements and release 2-3 free neutrons. “Prompt gamma rays” are produced concurrently with the free neutrons [9].

The second mechanism used as a neutron source is spallation. Spallation is a process where fragments of a material are ejected from a body due to stress or impact. A spallation source produces pulsed or quasi-continuous neutron beams through the acceleration of protons hitting a target material of a heavy nuclei and releasing neutrons. These heavy nuclei include lead, mercury, tantalum and etc. In contrast to the above mentioned mechanism, one spallation reaction releases about 20-30 neutrons per incident proton particle.

Neutron radiation is often referred to as indirect ionizing radiation. Unlike charged particles that ionize materials through electron excitation, neutrons have no charge and therefore cannot excite electrons. Despite their indirect ionization, neutrons are largely ionizing and this is best observed from the following example: when a neutron is absorbed and results into gamma ray emission, the gamma ray subsequently removes an electron from an atom or a nucleus recoiling from a neutron interaction is ionized and therefore causing more traditional subsequent ionization in other atoms. Since neutrons are uncharged, they are more penetrating than alpha and beta radiation. Neutron radiation from fission can be achieved through an effective fission chain reaction. The neutrons produced during fission should only be captured from a fissionable nuclei which then in turn splits and releases more neutrons [9].

What effects does neutron radiation have on materials? High-energy neutrons will not only degrade and damage materials but neutron bombardment to materials will create collision cascades. These cascades produce point defects and dislocations within the material which are responsible for the microstructural changes occurring over time to materials that are exposed to radiation. The radiation damage in the material occurs as a result of the interaction of an energetic particle with a lattice atom within a material. The collision causes a significantly large amount of kinetic energy transfer to the lattice atom from the neutron, the atom is then displaced from its lattice site and becomes what is known as a primary knock-on atom (PKA). As the PKA’s collide with each other, they lose energy with each collision and terminate as interstitials; and effectively creating a series of Frenkel defects within the lattice. Another consequence of the collisions is heat, this is created from the electronic energy loss. The magnitude of the damage caused by a single 1 MeV of a neutron creating a PKA in an ion lattice is such that it will produce approximately 1100 Frenkel pairs. This entire cascade of events happens over a very small time-scale of $10^{-13}$ seconds and can therefore not be observed without the aid of computer simulations [9].

3.2. The Monte-Carlo N-Particle 5 code
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 Monte Carlo N-Particle (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 [10]. The code takes into account the absorption and the moderation of the neutrons as they travel though matter.

Considering a beam of neutrons passing through bulk matter, the intensity of the beam will decrease as the beam travels through the material as neutrons are being removed by nuclear reactions. Slow thermal neutrons disappear through capture in the form of $(n, \gamma)$ reactions. The cross section of capture reactions are often dominated by one more resonances causing the
cross section to become very large. When the reactions are off resonance, the cross section decreases with increasing velocity with the relationship: \( v^{-1} \). The neutrons slow down and become moderated due to the elastic and inelastic scattering processes that occur as the cross section decreases therefore there is a higher probability of absorption to occur. The probability for resonant or non-resonant absorption to occur for neutrons is high in the eV energy range, this is the energy range neutrons are in after undergoing scattering processes from the initial energy of 1 MeV.

The loss in intensity of the neutron beam is given by:

\[
dI = -\sigma_t ndx
\]

(1)

where \( \sigma_t \) is the total cross section that includes the scattering process, \( ndx \) is the atoms per unit surface of the beam or material that the neutrons will encounter, \( n \) is the number of atoms per unit volume of the material and \( dx \) is the thickness of the material. The intensity will decrease with absorber thickness according to an exponential relationship given by:

\[
I = I_0 e^{\sigma_t nx}
\]

(2)

Considering an elastic collision between a neutron with initial energy \( E \) and velocity \( v \) with a target atom with mass \( A \) that is initially at rest, then applying the laws of conservation of energy and linear momentum will give the following ratio between the final neutron energy \( E' \) and initial energy:

\[
\frac{E'}{E} = \frac{A^2 + 1 + 2A \cos \theta}{(A + 1)^2}
\]

(3)

where \( \theta \) is the scattering angle in the centre-of-mass system. When there is no scattering occurring, the ratio will be given by \( E'/E = 1 \). The maximum energy loss occurs during a head on collision, when \( \theta = 180^\circ \) and can be expressed as:

\[
\left( \frac{E'}{E} \right)_{\text{min}} = \left( \frac{A - 1}{A + 1} \right)^2
\]

(4)

These expressions only apply for a single scattering event but since each neutron will scatter more than once, the energy loss has to be calculated repeatedly. The MCNP code does this calculation, a detailed discussion is found in S Krane [12].

The calculations can be made more quantitative by defining a parameter \( \xi \) to represent the average value of \( \log E'/E \) after a single collision, this parameter can be expressed as:

\[
\xi = \left[ \log \frac{E'}{E} \right]_{av}
\]

\[
= \frac{\int \left[ \frac{(A+1)^2}{A^2+1+2A \cos \theta} \right] d\Omega}{\int d\Omega}
\]

(5)

(6)

where \( d\Omega \) is the element of solid angle in the centre-of-mass system. Assuming that the scattering is isotropic then the integration can be carried out and give:

\[
\xi = 1 - \frac{(A - 1)^2}{2A} \log \frac{A - 1}{A + 1}
\]

(7)

The average value of \( \log E' \) is decreased after each collision by an amount the parameter represents and therefore after \( n \) collisions, the average value of \( \log E' \) will be \( \log E'_n \) and given by:

\[
\log E'_n \simeq \log E - n\xi
\]

(8)
This equation follows equation (5) above.

The calculations above are a good approximation for neutrons in the MeV energy range but as thermal temperatures are approached, the thermal motion of the atoms of the moderator are comparable to the speeds of the neutrons and the above calculations cannot account for their energy loss. A statistical mechanics approach to the problem will give a better approximation. Assuming that after sufficient time the neutrons will reach a thermal equilibrium with the moderator at a certain temperature then the neutrons can be described by a Maxwellian speed distribution given by:

$$f(v)dv = 4\pi n \left( \frac{m}{2\pi kT} \right)^{2/3} v^2 e^{-mv^2/2kT} dv$$

where \(f(v)dv\) gives the fraction of neutrons with speeds \(v\) and \(v + dv\), \(m\) is the mass of the neutron and \(n\) is the total number of neutrons per unit volume. The above expression can be written in terms of energy, giving [11]:

$$f(E)dE = \frac{2\pi n}{(\pi kT)^{3/2}} E^{1/2} e^{-E/kT} dE$$

4. Experimental procedure

The same experimental procedure followed in the proton irradiation study will be followed in this study too, with a few minor changes. The same plastic scintillator samples, polyvinyl toluene (pvt) and polystyrene (ps) based plastic scintillators purchased from ELJEN technologies and Saint Gobain Crystals will be studied. In addition to commercial scintillators, standard tile calorimeter plastic scintillators manufactured at CERN will also be studied. Instead of using the stopping range of ions in matter (SRIM) program to simulate the damage caused by the neutrons, the MCNP 5 code will be used to simulate the cascade event and calculate the energy loss and dose rate. The samples will be cut and polished in our sample preparation lab located within the Physics department. The samples will be irradiated with different dose rates of neutrons produced in favourable nuclear reactions using a radiofrequency linear particle accelerator as well as from the SAFARI nuclear reactor at NECSA and at the Joint Institute of Nuclear Research (JINR) in Dubna. Light transmission and light yield tests will be performed in order to assess the radiation damage on the scintillators. In addition, Raman spectroscopy and Electron Paramagnetic Resonance (EPR) analysis will be used to characterize the samples after irradiation and compare those results to un-irradiated samples.

5. Additional work

The project aims to extend these studies to include radiation assessment damage of any component that processes the scintillating light and deteriorates the quantum efficiency of the Tilecal detector, namely, photomultiplier tubes (PMT), wavelength shifting optical fibres and the readout electronics. The light that is emitted by the scintillator and passed through wavelength shifting optical fibres is detected by the PMT. The light that enters the PMT is detected and produces an output signal through a series of processes. The light passes through the input window to excite electrons in the photocathode such that photo-electrons are emitted into the vacuum. The photoelectrons are then accelerated and focused by focusing an electrode onto the first dynode where they are multiplied from secondary electron emission. The secondary electron emission is repeated at each of the successive dynodes. The multiplied secondary electrons that are emitted from the last dynode are finally collected by the anode [13].

PMTs will allow for a well conducted comparative study to determine which dynode material is more efficient and sustains the least damage from radiation. Major secondary emissive materials used for most dynodes are alkali antimode, beryllium oxide (BeO), magnesium oxide (MgO), gallium phosphide (GaP) and gallium arsenic phosphide (GaAsP), these will be some
of the material intended to be studied in the project [13]. The same neutron irradiation and damage assessment procedures followed for the plastic scintillators will apply to the dynode materials.

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