A comparative study of the high fluence neutron radiation effects on the properties of plastic scintillator UPS-923A for the TileCal of the ATLAS detector

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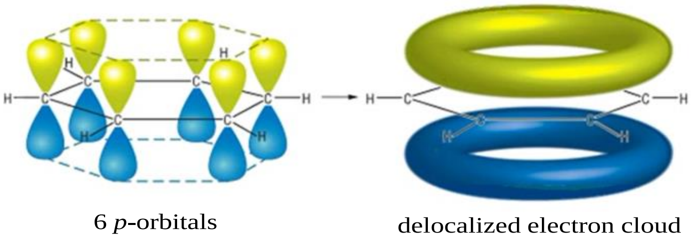
Abstract. The Ukrainian Polystyrene-based Scintillator (UPS-923A) doped with PTP and POPOP has been irradiated with a beam of neutrons. The neutron beams were provided by the IBR-2 pulsed reactor at the Frank Laboratory of Neutron Physics at the Joint Institute for Nuclear Research in Dubna, Russia. The neutron fluence ranged approximately between $10^{13}-10^{16}$ neutrons/cm^2. A yellowish to brownish coloration in the plastic scintillator samples was observed after irradiation. The radiation effects on the mechanical and scintillation properties were investigated by conducting Raman spectroscopy and fluorescence yield measurements. The results obtained show a substantial drop in scintillation intensity, of 65% loss after neutron bombardment. This effect is attributed to radiation-induced absorption of the fluorescence light by the damaged polystyrene base. A slight change in the Raman intensity of aromatic structures was also observed.

1. Introduction

Plastic scintillators are organic hydrocarbon compounds that are capable of emitting light when they interact with ionizing radiation [1]. They have many advantages such as fast rise and decay time, high optical transmission for enabling efficient data acquisition, as well as the fact that the manufacturing process is straightforward. On the other hand, inorganic crystals are more difficult to fabricate. Plastic scintillators are more cost effective when covering the typically large detector areas. For these reasons, plastic scintillators are widely used around the world for radiation detection in the scientific and industrial fields. The calibre of radiation detector depends primarily on the quality of the plastic scintillator employed.

The radiation detector systems used in high energy physics experiments such as the ATLAS detector of the Large Hadron Collider at CERN drives the need for use of plastic scintillators that can withstand the harsh radiation environment. This dire need directs attention to study radiation hardness of currently available plastic scintillators, to aid with the selection criteria of suitable fluorescence materials and ultimately develop radiation-hard scintillators for the future.

The plastic scintillator under investigation consists primarily of polystyrene base doped with 2% p-terphenyl (PTP) and 0.03% diphenyloxazolyl- benzene (POPOP) organic fluors [2]. The polystyrene base is a synthetic of aromatic benzene-ring structures that influence the scintillation properties of plastics. The chemical bonds present in a benzene ring are made up of σ-bonds which are in-plane and π-bonds which are out-of-plane of the benzene ring and overlap, as illustrated in Fig. 1. The π-bonds give rise to a delocalized electron clouds above and below the molecular plane as shown.



6 p-orbitals

delocalized electron cloud

Figure 1: Representation of chemical bonding in a benzene molecule.

The π-electron structure plays a key role in the scintillation mechanism of plastic scintillators during interaction with energetic particles. Incident radiation causes electronic excitations within the benzene molecule. The de-excitation of π-electrons to the ground state through fluorescence results in the emission of photons within the visible light spectrum [1]. The neutron radiation damage of plastic scintillators has been studied and reported in Refs. [3, 4, 5]. This study forms a comparative investigation effort in understanding radiation hardness of plastic scintillators at high neutron fluences.

2. Experimental details

The structural properties of radiation damaged polystyrene-based scintillator UPS-923A were investigated. The samples were cut and polished to dimensions of 14.4 x 7.2 cm with 6 mm thickness at the Institute for Scintillation Materials (ISMA, Kharkov, Ukraine). The irradiation of samples was performed at the Frank Laboratory of Neutron Physics (FLNP) at the Joint Institute for Nuclear Physics (JINR) in Dubna, Russia using the IBR-2 pulsed reactor [6, 7].

The samples were bombarded with a beam of neutrons provided by the IBR-2 reactor operated at an average power of 1875 kW. In order to achieve varying target neutron fluences, the samples were placed at different positions away from the reactor core and water moderator. In addition, during the irradiation only neutrons with energy $E > 1$ MeV were monitored since they are the ones which provided high neutron fluences. The neutron fluence ranged approximately between $10^{13}-10^{16}$ neutrons/cm^2. After irradiation, coloration of the samples was observed as neutron fluence increased. Figure 2 shows a photograph of the samples after irradiation.

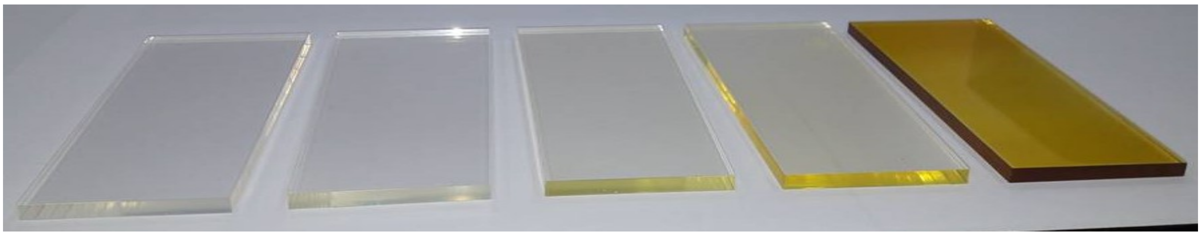


Figure 2: Neutron irradiated UPS-923A scintillators. From the left to right: non-irradiated, 10^{13}, 10^{14}, 10^{15} and 10^{16} neutrons/cm^2.

The assessment of structural disorder in samples after irradiation was performed by conducting Raman spectroscopy measurements. Raman spectroscopy is perhaps the most valuable method because it provides readily distinguishable structural signatures of carbon species present in the material. It is based on the Raman effect which is the result of inelastic scattering of light. The incident laser light interacts with lattice phonons leading to virtual excitations which then de-excite by releasing a photon of wavelength corresponding to a specific virtual mode. These Raman active modes are characteristic of certain bonding structures in the material. Raman spectra for the non-irradiated control samples as well as the irradiated samples were obtained using Horiba LabRAM HR Raman spectrometer. A 785 nm diode laser was used to excite the Raman modes and the spectrograph was calibrated via the zeroth order reflection of a white light source from the grating.

The scintillation efficiencies of irradiated samples were measured using the Horiba LabRAM HR Raman spectrometer. A laser excitation wavelength (λ_{ex}) of 244 nm, with a power of $\sim$20 mW at the source spot size of 0.7 μm was used to excite light emission through luminescence, and a laser spot size of 0.7 μm provided energy that allowed molecular excitations to occur. A grid of 11 x 11 points (121 acquisition spots) was mapped across a surface area of 200 x 200 μm using a motorised X-Y stage. This allowed for an average representative spectrum to be determined largely free from local variations introduced by surface features such as scratches.

3. Raman Spectroscopy Results and Analysis

Raman spectroscopy measurements were performed for non-irradiated control samples and irradiated samples. Figure 3 shows selected regions of Raman spectra containing aromatic structures. These structures are labeled as peak number 1 to 4 as shown in the figure. The peaks were assigned to their characteristic vibrational groups [8, 9].

c ring chains

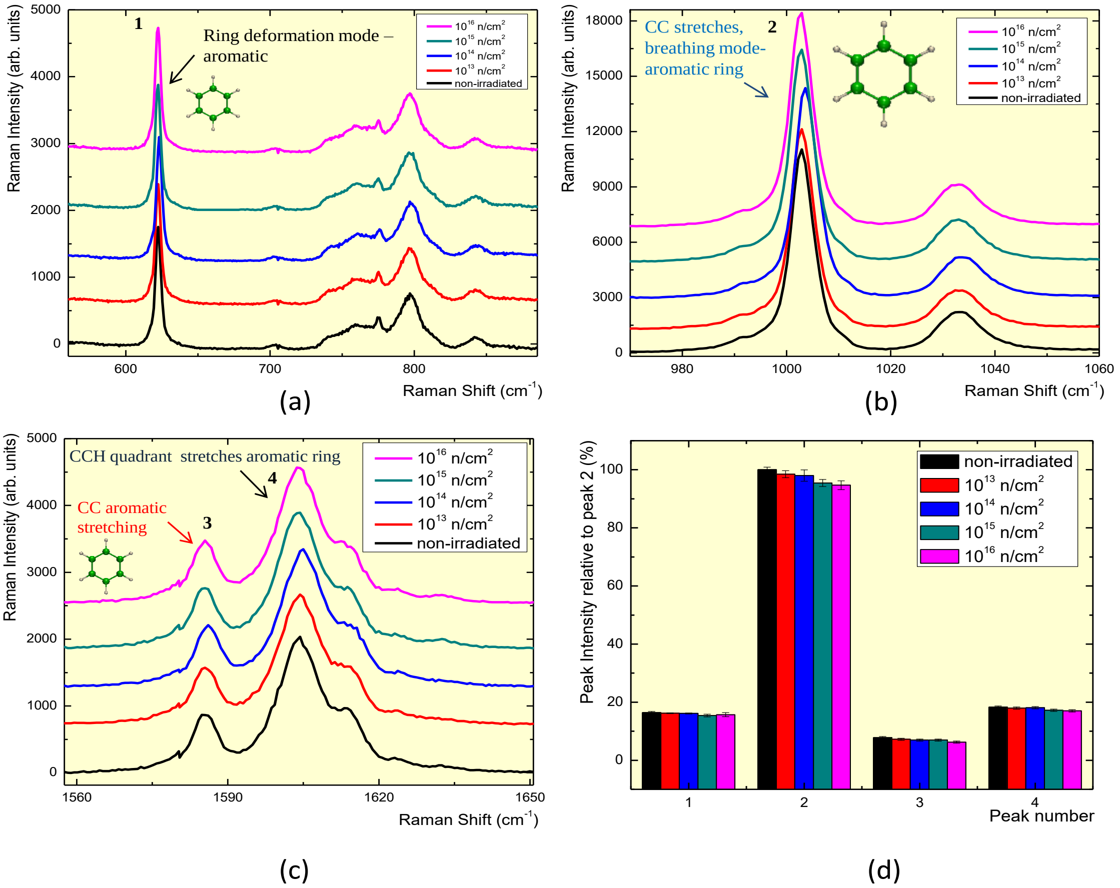
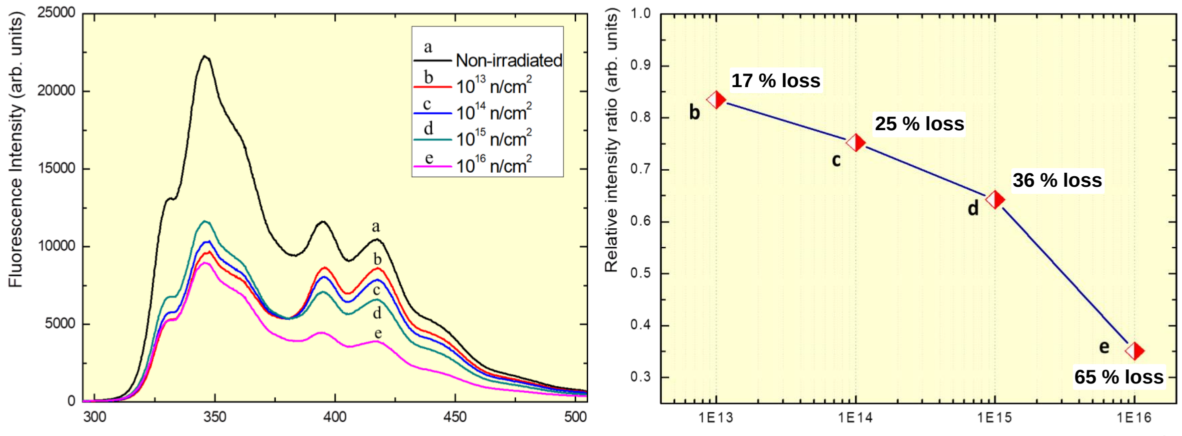


Figure 3: (a), (b) and (c)-Selected regions of background subtracted Raman spectra containing aromatic structures and (d)-Plot of intensities of peaks relative to peak 2 (non-irradiated) representing C–C breathing mode. NB: The Raman spectra have been vertically offset for better visual presentation.

The aromatic structures were assessed for radiation damage. The intensities of peaks were plotted relative to peak 2 (non-irradiated) in order to gauge change in the concentrations. Peak 2 corresponds to the breathing mode of the benzene ring. This is the most intense peak that largely influences the scintillation properties of plastics. With reference to Fig. 3(d), it can be observed that the intensities of peaks 1, 3 and 4 remain almost the same after irradiation. However, peak 2 becomes less intense as the neutron fluence increases. Torris, 2002 [10] ascribes this effect to a strong dehydrogenation due to the weak C–H bond breakage in the benzene ring and an emission of different C_XH_y groups with the absorbed neutron fluence. This reduces the concentration of species present in the material.

4. Fluorescence Spectroscopy Results and Analysis

Figure 4(a) shows the fluorescence spectra of non-irradiated and irradiated UPS-923A samples over a wavelength region of 300–505 nm. Several peaks are observed in the emission spectra. The wavelength regions at 300–375 nm and 375–505 nm correlate with fluorescence of the polystyrene base and organic fluor dopants, respectively. After irradiation of samples, a significant intensity drop is observed over the base region in comparison with fluor dopants region. There is competition for light absorption between the radiation-induced free radicals and fluor dopants [11].



(a) (b)

Figure 4: (a)-Fluorescence spectra and (b)-Relative intensity ratio as a function of neutron fluence measured at 430 nm wavelength, for UPS-923A samples.

Figure 4(b) shows relative intensity losses as a function of neutron fluence. This was measured at a wavelength of 430 nm since it correlates with the peak absorption wavelength of the wavelength shifting optical (WLS) fibers. An intensity loss of 65% is observed at higher neutron fluence of 10^{16} neutrons/cm^2, compared with much lesser percentage losses at lower fluences.

5. Conclusion

The structural and scintillation properties of Ukrainian polystyrene-based UPS-923A scintillator irradiated with a beam of neutrons were investigated. All the irradiated scintillator samples reveal evidence of radiation damage. A yellowish to brownish coloration in the plastic scintillator samples was observed after irradiation with higher neutron fluence. When a plastic scintillator is bombarded with high energy particles, weak C–H chemical bonds within the material are broken and form free radicals. The free radicals initiate chemical reactions and form absorption color centers which could account for the observed coloration [10].

The aromatic benzene structures present in the scintillator material were identified and assessed through Raman spectroscopy. These species play a very important role in the scintillation mechanism since they give rise to delocalized electron clouds responsible for fluorescing upon excitation by incident ionising radiation. Amongst the four aromatic structures identified (peak 1 to 4), a noticeable slight intensity decrease was only observed in peak 2. This peak largely contributes to the scintillation yield of plastics, it typically represents C–C ring breathing mode. The decrease in the intensity is ascribed to breakage of chemical bonds after irradiation.

From the fluorescence results obtained, it is evident that neutron bombardment affects the luminescence properties of plastic scintillators. A significant drop in the fluorescence yield is observed. At higher neutron fluences, radiation induced free radicals absorb the fluorescence light that could have been detected. The damage to the π-electron structure within the polymer matrix also accounts for the fluorescence yield losses.

6. Acknowledgement

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