In-situ characterization of radiation hardness of scintillating targets using heavy ion beams

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Abstract. Scintillating materials are of interest for numerous applications related to ionizing radiation. Exposure to radiation induces damage in these materials, resulting in degradation of their scintillation properties. The knowledge of radiation damage rate as a function of dose is especially important when scintillators are exposed to heavy-ion beams. In this contribution, we report the development of a setup for in-situ characterization of radiation damage of scintillation targets exposed to heavy-ion beams. This setup provides a unique opportunity to study scintillating properties of materials under heavy ion exposure with energies of 300 MeV/u and above. The description of the setup and the sample characterization procedure are discussed on the basis of a study of zinc oxide ceramic scintillators.

1 Introduction

Materials that exhibit light emission when excited by ionizing radiation are called scintillators. These materials make it possible to detect ionizing radiation, such as electrons, protons, gamma-rays, alpha-particles, ions, etc. One can use scintillators to measure radiation intensity, beam position and beam distribution in space and time, by measuring the characteristics of the emitted light. Scintillators are found in industrial applications and also used for many scientific research purposes worldwide (mainly dealing with X-ray, or gamma-ray radiation).

At the GSI Helmholtz Centre for Heavy Ion Research scintillating materials are used as active material in detectors for heavy-ion accelerator beam diagnostics. In this case, scintillators are exposed to various ion species from hydrogen to uranium, that can be accelerated with the GSI-FAIR accelerator complex [1, 2]. The radiation doses deposited by heavy ions (e.g., uranium) in scintillating materials are orders of magnitude larger than for the most common X-ray and gamma-ray irradiation applications. Such extreme radiation conditions can cause material damages that change scintillating properties which are critical for operation of certain devices.

In this contribution, we describe a setup we built for in-situ characterization of the change of scintillation light intensity and wavelength spectrum as a result of extended exposure to high-energy heavy-ion radiation of few hundreds MeV/u. With this setup experimental data for comparison of radiation hardness of plastic scintillators (BC400) and of inorganic ceramic scintillators based on zinc oxide doped with indium or gallium is collected.

2 Experimental setup

The conceptual layout of the measurement setup is shown in Figure 1. A scintillating target, in the figure named “sample”, is directly inserted into an ion beam. The beam energy is sufficiently high to pass through the scintillating target and exit from the opposite side. The scintillation light is emitted from both surfaces when the ion is entering and exiting the scintillating target and light intensity, spectrum, and the beam spot position are monitored by means of different photosensitive devices (digital camera, optical UV/Vis spectrometer and photomultiplier tubes).

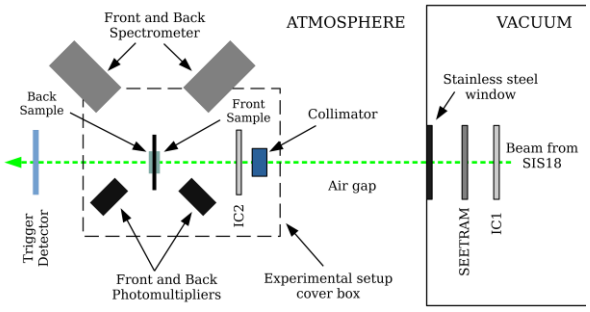

Fig. 1. Conceptual scheme of the in-situ scintillation light measurement. The scheme is taken from [3].

The incident beam intensity and the accumulated radiation dose are monitored by means of secondary electron transmission monitor (SEETRAM) and ionization chambers (IC1 and IC2) placed prior to the investigated target (see Figure 1). In this work, we use ion fluence (integral number of incident ions per unit area) as a measure of radiation dose.

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The photo of the experimental setup for in-situ characterization of scintillating targets is shown in Figure 2.

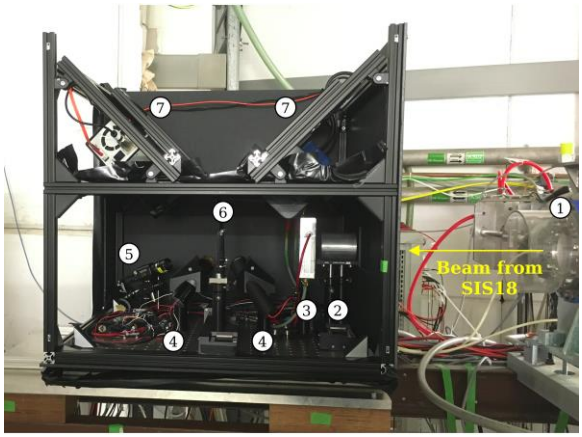

Fig. 2. Photo of the experimental setup for the in-situ characterization of scintillating targets exposed to high energy heavy ion beams [3].

In Figure 2, the black box of the setup is partially opened to show the equipment inside:

1 – vacuum window separating the beam line from atmosphere,
2 – beam collimator is used to define 5 mm diameter beam spot,
3 – ionization chamber for measurement of the incident beam intensity and the ion fluence,
4 – photomultiplier tubes (PMT) for scintillation light intensity measurements,
5 – digital camera for imaging and for beam adjustments on target,
6 – target ladder on a stepper motor stage to select different targets for irradiation,
7 – spectrometers for scintillation light spectrum measurement.

A photo of the target ladder is shown in Figure 3. Due to the limited access to the experimental area, multiple targets are mounted on the ladder so that the position to be irradiated can be selected remotely via the motorized stage. In addition to investigated samples, a CROMOX (Cr-doped Al_2O_3) fluorescence screen is used for beam position and focusing prior to the target irradiation.

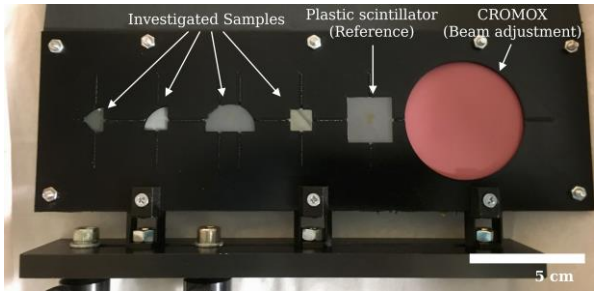

Fig. 3. Photo of the target ladder with the investigated scintillating targets [3].

2.1 Scintillation light intensity measurement

The total intensity and time evolution of the emitted scintillation light are measured by means of Hamamatsu H7415 and H13661 PMT assemblies, respectively. The key parameters of these assemblies are summarized in Table 1.

Table 1. Basic properties of the PMT used in the experimental setup.

Property	H7415	H13661
Spectral response	300 to 650 nm	
Maximum sensitivity	420 nm	
Rise time (typical)	1.7 ns	0.23 ns
Gain	$5x10^6$	$2x10^4$

Signals from PMTs were recorded using a Tektronix MSO 5B 2 GHz oscilloscope. The scintillation light intensity was estimated using signals of the H7415 PMT. The H13661 was used for the time-resolved scintillation intensity measurements. Each PMT signal corresponds to a single ion impact into the scintillating target. The PMT signal recording was triggered by the trigger detector inserted into the beam further downstream behind the setup, as shown in Figure 1. The average intensity of scintillation light (light yield) emitted per single ion impact in the scintillating target as well as the time-resolved characteristics (e.g., scintillation rise and decay time) are evaluated from approximately 50000 signals.

For the light yield, as well as rise and decay time measurements as a function of fluence the ion beam intensity was set to 10^7 – 10^8 ions/s for fluence accumulation. For recording the PMT signals the intensity was lowered to 10^3 – 10^4 ions/s to reduce the number of signal pile-up events when two ions hit the target within too short time one after another.

2.2 Scintillation light spectra measurement

The scintillation light spectra are collected by means of spectrometer assemblies as shown in Figure 4. The spectrometers are mounted on the top of the setup (see Figure 2). The first assembly registers the light emitted towards the front side of the setup and the second registers the light emitted towards the rear side of the setup. The use of two spectrometers makes it possible to characterize simultaneously two scintillating targets stacked together with a piece of black paper in-between.

Each spectrometer assembly consists of two parts: 1) a spectrograph which splits the incoming light into a spectrum, and 2) a digital camera which records the output spectrum in a digital format. For both spectrometers a Horiba CP140-202 spectrograph is used, which is capable of covering a spectral range of 190 – 800 nm with an average dispersion of 50 nm/mm.

For the spectrum recording the digital camera PCO.1600 was applied in the front and a PCO SensiCam QE in the rear assembly. Both cameras are equipped with an external trigger capability. Therefore, the spectra acquisition can be synchronised well with the incident pulsed ion beam. The PCO SensiCam QE has higher sensitivity in the UV region than the PCO.1600. More specification details can be found on the PCO web page [4, 5].

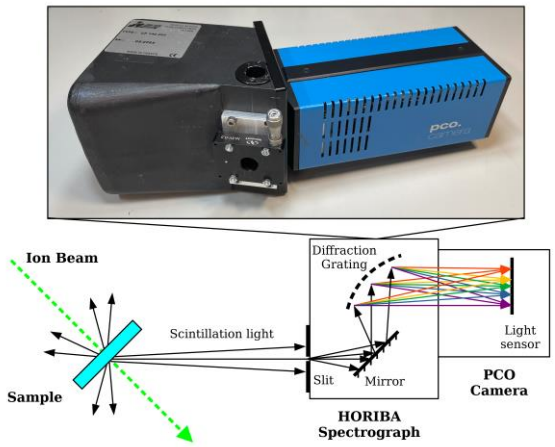

Fig. 4. Photo of the spectrometer assembly and a schematic view of the scintillation light path through the spectrometer assembly [3].

Heavy-ion beams from the SIS18 synchrotron are delivered to the experimental setup as spills. Each spectrum acquisition corresponds to an individual spill. The spectrum acquisition time varies between 1 and 5 seconds depending on the spill duration and number of ions in the spill. The number of ions per spill that hit the scintillating target is measured using the ionization chambers IC1, IC2, and secondary electrons transmission monitor SEETRAM (see Figure 2). This number is used later for normalization of the recorded spectra intensities.

Spectra are acquired concurrently with the accumulation of fluence in the investigated scintillating target. Achieving high fluence requires high beam intensities. However, some scintillating targets are highly sensitive to intense heavy-ion radiation, which can cause significant radiation damage already during the first spectrum acquisition (this measurement is later considered to represent the pristine state of the sample). Therefore, selecting the appropriate beam intensity involves a trade-off: higher intensities improve spectral signal quality and allow faster fluence accumulation, while lower intensities help minimize radiation damage during spectrum acquisition. In our experiments, optimal conditions were achieved using approximately 10^7 to 10^8 ions per spectrum acquisition.

3 Investigated targets example

We investigated different zinc oxide based scintillating targets as an alternative to the BC400 plastic scintillator. These ceramic scintillator samples were produced at the JSC Vavilov State Optical Institute, St. Petersburg, Russia. The samples were prepared by uni-axial hot-pressing in vacuum. Zinc oxide powder was mixed with indium oxide or gallium oxide powder to reach a doping concentration of about 0.1 wt.%. Due to trade secrets of the JSC Vavilov State Optical Institute not all details of sample preparation can be disclosed. More details on the ceramics preparation and material properties can be found elsewhere [6-8].

For the high-energy heavy-ion irradiation tests, the ceramic scintillator samples were cut to thicknesses of 0.5 to 1 mm and a size of 0.25 to 4 cm^2. Figure 5 shows selected photos of the investigated scintillating targets.

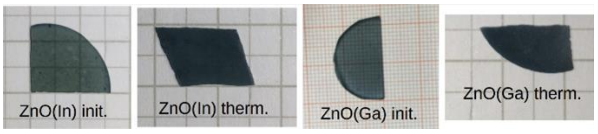

Fig. 5. Selected photos of the investigated scintillating targets made of ZnO-based ceramics.

4 Spectroscopy measurement results

4.1 Scintillation light spectra

Figure 6 shows the change in scintillation light spectrum of the indium-doped zinc oxide ceramics, ZnO(In), exposed to a ^{209}Bi beam at 300 MeV/u energy. One can see a single emission band with a maximum intensity at 385 nm wavelength. This band has been addressed in the earlier publications as near-band-edge emission (NBE) and is known to have ultra-fast scintillation decay times less than 1 ns [6-10].

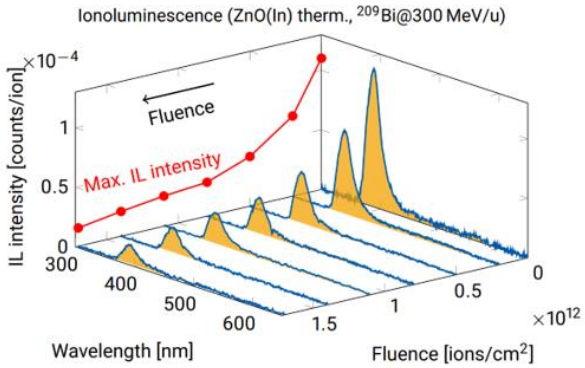

Fig. 6. Scintillation light spectra as a function of fluence of ZnO(In) ceramic target exposed to ^{209}Bi ion beam at 300 MeV/u energy [3].

The highest intensity of the emitted light is observed for the pristine sample. Then the intensity drops with increasing fluence due to the accumulation of radiation damage. It should be noted that for the ZnO(In) sample no new emission peaks are formed as the radiation damage is created. This indicates that the material remains a fast scintillator without any afterglow occurring as a result of ion irradiation.

4.2 Critical fluence evaluation

In order to perform a quantitative comparison of different scintillation materials with respect to their

radiation hardness we use the Birks-Black model to extract the so-called critical fluence value [11]. According to this model, the intensity of scintillation light as a function of fluence can be written as:

$$I(\Phi) = I_0 / (1 + \Phi/\Phi_{1/2}),$$

where I_0 is the initial intensity of the scintillation light of the pristine sample, Φ is the fluence, and $\Phi_{1/2}$ is the critical fluence. One can interpret the critical fluence as the fluence value at which the scintillation light intensity drops by 50% of the initial intensity.

Figure 7 shows the critical fluence values obtained after targets exposure to different ion species. Therefore, critical fluence is studied as a function of the stopping power of high energy heavy ions in material [3]. Using the experimental setup presented here, it has been demonstrated that zinc oxide-based ceramics can be considered as a promising alternative material to plastic scintillators. ZnO(In) has orders of magnitude higher critical fluence compared to BC400 plastic scintillators. Therefore, heavy ion beam diagnostic's detectors built from zinc oxide have the potential of longer operation time significantly postponing the replacement necessary due to damages at the scintillation detector.

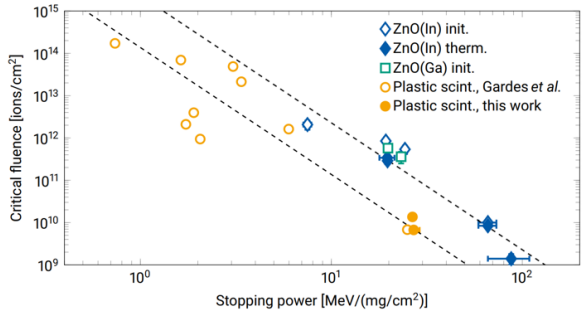

Fig. 7. Critical fluence as a function of stopping power for ZnO(In) and BC400 scintillators taken from [3].

5 Summary

We designed and built a customised setup for in-situ characterization of various scintillating targets under high-energy heavy-ion beam exposure. The setup is capable of measuring scintillation light yield and wavelength spectrum change as a function of ion fluence. This setup provides a unique opportunity to study scintillation properties of materials under irradiation with heavy ions with energies of 300 MeV/u and higher. The high energy leads to uniform energy distribution in the investigated sample.

The setup enabled us to compare the radiation hardness of ZnO-based ceramics and BC400 plastic scintillators and helped to test and select promising materials for a further upgrade of scintillation detectors operating in heavy-ion radiation conditions.

Acknowledgments

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