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High resolution charge spectroscopy of heavy ions with FNTD technology

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ABSTRACT

This paper is focused on the improvement of the heavy charge particle charge resolution of Fluorescent Nuclear Track Detector (FNTD) technology. Fluorescent intensity of individual heavy charge particle tracks is used to construct the spectrum. Sources of spectroscopic line broadening were investigated and several fluorescent intensity correction procedures were introduced to improve the charge resolution down to $\delta Z = 0.25$ c.u. and enable FNTD technology to distinguish between all projectile fragments of 290 MeV carbon ions. The benefits of using FNTD technology for fragmentation study include large dynamic range and wide angular acceptance. While we describe these developments in the context of fragmentation studies, the same techniques are readily extended to FNTD LET spectroscopy in general.

1. Introduction

Nuclear fragmentation measurement data of heavy ions propagating through matter provides a rich source of information not only for fundamental studies on nuclear reaction models [1–3] but also are important for practical applications such as heavy ion radiotherapy [4,5] and the design of shielding from space radiation exposure [6]. Carbon ions with several hundred MeV/n energy have been successfully employed for cancer radiotherapy due to their clinical advantages including high relative biological effectiveness (RBE) and low oxygen enhancement ratio (OER) compared with conventional radiation modalities using photons and protons (e.g., [7,8]). The carbon-ion beam therapy takes advantage of the steep dose gradient in a Bragg-curve energy deposition profile in the patient body and eventually deposits highly conformed and precise dose in the tumor volume. Thus, unique dose distribution during heavy ion therapy provides the low radiation exposure of healthy tissue and adjacent critical organs while demonstrating effective therapeutic performance for tumors. However, the carbon ion beam generates secondary particles (fragments) produced by the nuclear fragmentation reactions while passing through the body, which would contribute to the unexpected dose exposure in healthy tissue and secondary cancer risk. Several experiments to measure charge-changing cross sections of carbon ions in the tissue equivalent materials like water have been carried out to estimate of those fragments dose contribution [9–13]. So far obtained data are still limited in the energy and nuclear charge range of fragments and exhibit large uncertainties, especially in the low energy region below 100 MeV/n near the Bragg-peak.

Charge-changing cross-sections have been measured with silicon active telescopes [12], CR-39 plastic nuclear track detectors (PNTDs) [10] and nuclear emulsion chambers [13]. Previous cross-section measurements were performed with $\delta Z < 0.2$ charge units (c.u.) with the use of CR-39 PNTD (0.1–0.2 c.u.) [10,14–16], silicon semiconductor detectors (0.1–0.2 c.u.) [17,18], and scintillation counters (0.13–0.14 c.u.) [19]. Silicon active telescopes yield a precise fragment charge spectrum with high statistical data but have limited angular acceptance. Multi-fragment production such as $C \rightarrow 3\alpha$ is difficult to detect as well as it is impossible to perform the measurement of emission angle of each fragment [20–23]. On the other hand, since passive detectors such as CR-39 and nuclear emulsion record fragment tracks, the fragment passage through the material can be easily reconstructed by connecting reaction vertices [13,22]. However, there are issues concerning the detection threshold and charge identification capability of those passive detectors. CR-39 has excellent charge and mass resolutions of

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~0.2 c.u. (amu) [24–27], however, CR-39 cannot detect proton or helium tracks at the required energies [28]. In contrast, nuclear emulsions register any particle with $Z \geqslant 1$, but the charge distribution for heavy ions greater than lithium ($Z \geqslant 3$) are excessively broadened making it difficult to identify individual nuclear charges without special techniques including a refreshment chemical process and multilayer averaging method [13,29]. The hybrid method which combines CR-39 and nuclear emulsion has been proposed for covering whole nuclear charge range of produced fragments [29,30]. However, obtaining trustworthy results with the hybrid method presents difficulty as the two techniques require distinct handling, chemical processing and readout procedures.

Fluorescent Nuclear Track Detector (FNTD) technology, based on the Al_2O_3:C,Mg single crystals, provides high sensitivity and high detection efficiency, not only for heavy charged particle but also for delta electrons [31–34]. Radiochromic transformations of color centers occur in the high-ionization-density core of a track within the crystal creating a "fluorescent track" analogous to those recorded in CR-39 and nuclear emulsions. FNTDs enable diffraction-limited optical imaging of ionization patterns as readout is performed with fluorescence laser scanning confocal microscopy (LSCM). Therefore, the FNTD method is all-optical, does not require any chemical processing and allows for easy automation, 3D imaging and subsequent 3D image processing. FNTDs are not sensitive to ambient room light before, during or after the irradiation and allow for nondestructive imaging and re-readability. The short fluorescent lifetime of constituent color centers which undergo radiochromic transformation allows for the use of fast, high-productivity imaging. Furthermore, the saturation level of track density is many times higher than that for other track detectors due to the small, diffraction-limited size of the imaged tracks. FNTDs exhibit extremely wide dynamic range for heavy ion detection from proton ($Z = 1$) to xenon ($Z = 54$) without complete saturation of fluorescent amplitude [35,36]. Therefore, FNTDs allow for the measurement of the nuclear charges of fragments in a single detector, without the need for combination of CR-39 and nuclear emulsions. Charge spectroscopy with FNTD technology, up to this study, has yielded broader charge distributions than CR-39, though narrower than nuclear emulsions [35].

In this paper, we demonstrate a significant improvement of the charge resolution of FNTDs by determining and reducing uncertainties which contribute to the previously poor charge and LET resolution [36]. Several sources of broadening were identified including non-uniformity of color center concentration within FNTD crystals, the variation in energy deposition along heavy ion tracks and necessary corrections associated with the optical system of the FNTD reader.

2. Methods

To quantify the degree to which fragmentation peaks can be resolved we use two metrics: charge resolution and the coefficient of variation. Charge resolution, δZ, is calculated by examining the efficacy with which a technique distinguishes particles with charge Z from particles with charge Z-1. It is calculated by Eq. (1) as:

$$\delta Z = \frac{\delta A_z}{A_z - A_{z-1}} \tag{1}$$

Here, A represents the parameter which is a measure of ionization density. In FNTD technology, A represents the maximum fluorescence intensity measured from an imaged track. δA_z in this case is a measure of the spectroscopic line width corresponding to the distribution of fluorescent intensity values A of a particular charge species with atomic number Z.

An alternate method by which spectroscopic resolution can be estimated involves fitting the fluorescent intensity frequency distribution to a Gaussian function. One standard deviation of A values (σ parameter of a Gaussian function) divided by the average value of the fluorescent peak intensity (A_0) yields a measure of the peak width in percentage and is further referred to as the coefficient of variation (CV). Calculation of the CV is performed as:

$$CV = \frac{\sigma}{A_0}\% \tag{2}$$

Both the δZ and the CV are provided in this work for easy comparison.

2.1. Materials

FNTDs are single crystal plates polished on one side and made out of carbon- and magnesium-doped aluminum oxide (Al_2O_3:C,Mg). The main radiation sensitive fluorescent centers in this material are F_2^{2+}(2Mg) color centers which are aggregate defects consisting of two oxygen vacancies charge compensated by two magnesium ions [37]. During irradiation these color centers undergo radiochromic transformation by trapping electrons, but at the same time they are optically very stable so they are insensitive to room light and thermally stable up to 600 °C [37]. Optical bleaching with pulsed UV light provides a means to erase and re-use FNTD crystals. Optical UV bleaching also reduces the non-radiation related background signal, makes it more uniform across the acquired image and this way allows to detect charge particle tracks with LET as low as 0.5 keV/um.

2.2. Irradiations

The detectors were cut from a single Al_2O_3:C,Mg crystal in the form of $4 \times 6 \times 0.5$ mm^3 plates and were polished on one side. The detectors were exposed to carbon ($Z = 6$) ions at normal incidence with a nominal energy of 290 MeV/n produced by the Heavy Ion Medical Accelerator (HIMAC) at National Institute of Radiological Sciences (NIRS) in Chiba, Japan. A poly-methyl-methacrylate (PMMA) absorbers with different water-equivalent thicknesses (63.26 and 117.24 mm) were placed in front of the FNTDs to produce lighter fragments from primary Carbon ions. The irradiated beam fluence was approximately 10^6 cm^{-2}.

2.3. Detector readout

Images of irradiated FNTD crystals in fluorescent contrast were acquired using Landauer custom-built confocal laser scanning fluorescent imaging system (FNTD reader). For light excitation, this system employs a Blue Sky fiber-optically coupled 639 nm single mode laser diode with collimator providing 6 mm diameter beam. Thorlabs Laser Driver Controller 202C provided an 8.66 mW of laser power on a sample at 65 mA of laser diode current. 2D scanning galvanometers imaged with relay lenses on the back aperture of the 0.95 NA, 40× Nikon Plan Apo microscope objective provided a raster scan with typical 100 by 100 μm^2 field of view and 512 by 512 pixel image size. The 200 μm confocal pinhole equal to 2 Airy disk diameters provided adequate special resolution without degrading the sensitivity to low Z and linear energy transfer (LET) particles. Fluorescence was detected with a customized, 250 kHz Hamamatsu C10508 avalanche photo-diode (APD). Each 100 μm × 100 μm image was acquired in 10 s. Though some of the parameters were varied throughout this study, the final spectra were obtained using stacks of 40 images each separated by increment in depth $\Delta d = 3.0$ μm. A total of 400 stacks were acquired summing to a total area of 4 mm^2.