LiF Crystal probed by Ion Beam Induced Luminescence (IBIL) Analysis in Order to Identify a Linear Relationship Between Peak Intensities and Ion Beam Dose

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INTRODUCTION

Pure LiF crystals probed by ion beams give rise to the formation of well assessed color centers [1] characterized by visible fluorescence features at room temperature, whose light yield depends on the radiation dose. However, the comprehension of the behavior of luminescence bands under ion irradiation is still not straightforward. Moreover, this topic is actually of paramount importance because these materials could be used in hadrotherapy treatments [2] to monitor the dose of the ion probe during the cures.

Due to this, we analyzed pure LiF crystal by means of Ion Beam Induced Luminescence (IBIL) analysis in order to characterize the evolution of the luminescence optical features and to demonstrate that a linear relationship between the IBIL yield and the released dose can be obtained. Specifically, we used a multiple linear regression (MLR) method to identify a relation between the dose and the intensity values at selected wavelengths.

EXPERIMENTAL

IBIL measurements were performed by irradiating commercial nominally pure LiF crystals ($10\times10\times1$ mm^3, polished on both sides) at room temperature with a 2.0 MeV proton beam, perpendicular to the sample surface. The beam cross section was about 1 mm^2 and the current density was around 2.3 μA/cm^2, corresponding to an ion flux of around 1.4×10^{13} H$^+$/(cm^2 s). During the ion irradiation, performed at 10^{-6} Torr, the chamber worked as a Faraday cup allowing to measure both the total charge impinging on the sample and the beam current. The maximum reached fluence was 2.9×10^{15} H$^+$/cm^2. From the average proton range R_p = 37.9 μm in LiF calculated by SRIM2008 [3] taking into account the material density ρ = 2.635 g/cm^3, the fluence was converted into dose, reaching a maximum of 92 MGy. The ionization energy of the impinging protons ranges from 33 eV/(ion nm) at the sample surface up to a maximum of 120 eV/(ion nm) at the Bragg peak position. The luminescence light was collected into the vacuum chamber by a silica fiber (600 μm diameter) put in front of the irradiated surface, at a distance of around 15 cm and at an angle of 20° with respect to the beam line direction. IBIL spectra were recorded by an Ocean Optics QE65000 spectrometer coupled to the chamber by means of a fiber vacuum connector. During the irradiation, 200 spectra were recorded, each one within 1 s of integration time, allowing to follow the light intensity as a function of fluence within a wavelength interval ranging from 250 nm to 900 nm.

RESULTS

In Fig. 1 are shown the IBIL spectra of a LiF crystal, collected at 1.4, 51 and 93 MGy of irradiation dose, while in the 2-D plot of Fig. 2, the normalized intensity evolution of the full spectrum during the irradiation is represented (for each wavelength the intensities are normalized to its own maximum value). It is evident that each wavelength is characterized by a different and non linear trend and, at high doses, all the intensities reach a saturation value characterized by a low sensitivity. In order to analyze, at least from a conceptual point of view, the possibility of extracting a useful information on the irradiation dose from the IBIL spectra, we performed a MLR analysis of the luminescence intensities at different wavelengths, with the aid of the code StatSoft 8.0. In particular, we fit the dose values D with a multiple linear function of the type where β_i are the fitting parameters and I_i are the intensities at the λ_i peak wavelengths, as shown in equation 1.

$$D = \beta_0 + \sum_{i=1}^{N} \beta_i I_i; \tag{1}$$

With the intensity values at wavelengths near the main peaks, namely at 312 nm, 383 nm, 537 nm, 664 nm and 855 nm, the MLR was developed by means of a forward stepwise method. The best results were obtained by interpolating the intensities values at 312, 664 and 855 nm (R^2 value up to 0.99). In Table 1 are reported the MLR coefficients calculated for these three wavelengths. Due to the decrease of the luminescence yield during irradiation, all the values are negative.

Table 1. MLR regression coefficients and the corresponding standard errors.

	β Regression Coefficient	β Standard Error
Intercept	20.0	0.3
312 nm	-58.4	1.2
664 nm	-4.1	0.2
855 nm	-23.9	0.8

In Fig. 3 are shown both the experimental dose points and the final interpolation line as a function of the predicted values, calculated with the described MLR analysis. As can be observed, the experimental points are

very close to the line within the whole dose interval. It is worth noting that a very good interpolation was obtained with only three variables, allowing to realize a reliable calibration curve with a low waste of time and calculations.
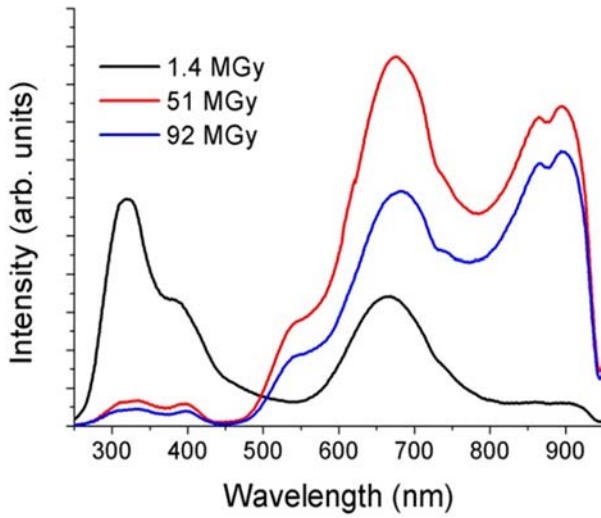

Fig. 1. IBIL spectra of the 2 MeV proton irradiated LiF crystal collected at 1.4, 51 and 92 MGy of irradiation dose.
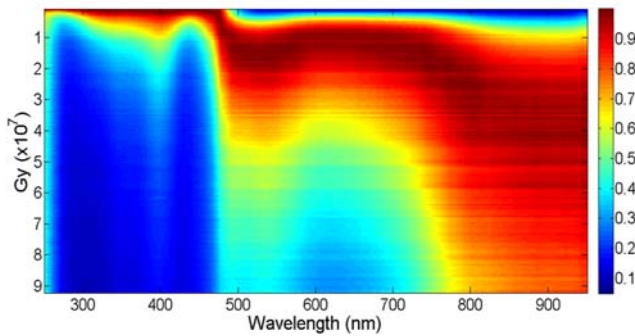

Fig. 2. Normalized intensities of IBIL spectra of the 2 MeV proton irradiated LiF crystal as a function of the dose and of the wavelength.
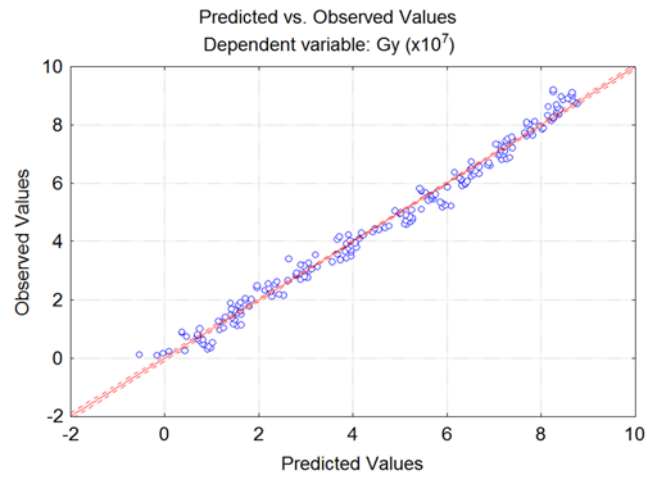

Fig. 3 Observed vs. predicted dose values obtained from the MLR of the luminescence yield at 312 nm, 664 nm and 855 nm. Circles are the experimental points and the red line is the interpolation curve obtained from MLR. Dashed lines indicate the 95% confidence interval for the interpolation curve.

A standard error of around 3 MGy was evaluated in the estimation of the predicted dose value from the MLR curve. The standard error is also related to the beam current fluctuations and could be improved by providing a set-up for recording the charge corresponding to every single spectrum.

It has to be pointed out that the investigated dose interval is quite large. For more practical applications the MLR analysis can be performed on narrower dose intervals, by definitely improving the signal to noise ratio.

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