IRSTI 29.15.39; 29.03.31

Dosimetry of Charged Particles Using Lyoluminescence Method

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Interaction of charged particles – protons, nitrogen, oxygen and carbon ions with LiF single crystals were studied using lyoluminescence method. Obtained linear energy transfer profiles were compared with those ones calculated by Monte Carlo method. The shape of the calculated and measured profiles was found to be comparable with Brag's curve. Analyses of differences in shape of calculated and experimentally measured curves were performed. It is demonstrated that by gradually dissolving the crystal in the form of a disk, rotating it with constant angular rate around its axis of symmetry, it is possible to determinate the defect distribution over the crystal depth with a resolution of the order of 1μm according to measurements of lyoluminescence intensity. Also, a good precision of lyoluminescence intensity measurement and agreement of experimental energy loss profiles with the theoretical ones are noted. Significant deviations from calculated profiles were observed in distances closer than 10^{-6} m to solid surface.

Key words: Lithium fluoride, lyoluminescence, dissolution, energy transfer, radiation defects, accelerated ions.
PACS number(s): 61.80.−x

1 Introduction

Defect's creation by accelerated ions in alkali halides has been extensively studied. These defects may be used as detectors of the particle-matter interactions and particularly of the energy loss processes [1]. Most of the created defects are well known and they can be easily revealed by optical methods, such as absorption and emission. For high-energy ions (E>MeV/amu) it is possible to measure the depth distribution profiles of primary point defects (F centres) and aggregated defects (F_2 centres) using microspectrophotometric technique [1-3]. For low-energy ions (E<MeV/amu) using of this method is problematic taking into account the small penetration depths (less than1 μm) of these ions. Lyoluminescence (LL) studies seem to be helpful aid for measurement of F centres distribution in collision-cascade volume produced by low-energy ions interaction with alkali halides [4]. Rotating disc technique allows using LL kinetic measurements under specified hydrodynamic condition (dissolution of LiF in sulphuric acid) to obtain information about distribution of point defects in irradiated solids [5, 6].

The present work has been undertaken to examine possibilities of the LL method for the determination of defects distribution over the crystal depth formed by bombardment with accelerated ions. As it was pointed out in [4], the LL light emission is very sensitive to the heterogeneous solvent – acid substance interactions and deviations from theoretical LL profiles in distances closer than 20 μm to surface. On the other hand, in case of additively coloured KCl single crystal such deviations have not been found [5]. To explain experimental results concerning the LL light emission, we have compared the LL intensity profiles to the calculated electronic energy loss profiles. The main goal of this short paper is to claim out rather good precision of LL intensity measurement and good agreement of experimental energy loss profiles with the theoretical ones. Significant deviations from calculated profiles were observed in distances closer than 10^{-6} m to solid surface. This range is ten times smaller than it was proposed earlier.

2 Experimental procedure

LiF (lithium fluoride) single crystal cylinders of 5 mm radius and 10 mm height were exposed to bombardment with H^+, He^+, N^+, C^+, and O^+ ions (see table 1). The irradiation was performed at room temperature using the beam line facility HIT of the Nuclear Investigations Centre of Tokyo University.

After irradiation an optical absorption analysis of the defects present in the crystal was done using a UV/VIS spectrophotometer. The shape of observed spectra was similar to those reported in [2, 7, 8]. The dominant colour centres in LiF irradiated at room temperature with accelerated ions have been identified previously [1-3, 7,8].

Dissolution of irradiated specimens was carried out using the rotating disc technique [4-6], $3\cdot10^{-4}$ M ruthenium bipyridyl chloride in 3M sulphuric acid aqueous solution was used as a solvent. LL intensity was measured using a unit specified in [9] at constant dissolution rate. As it was verified previously [5], LL intensity is directly proportional to the radiation damage. During dissolution under specified hydrodynamic conditions (at constant dissolution rate) LL intensity represents local concentration of radiation defects in thin layer dissolving at this moment of time [4, 5]. This feature provides a possibility to obtain the distribution profiles of the defects along the path of the incident ions. The thickness of the dissolved disk part is proportional to the time of dissolution [5]. Obtained linear energy transfer profiles were compared with those ones calculated by Monte Carlo method. The energy deposition curves were calculated using TRIM code Version 91.00 (original release TRIM-85 by Pergamon Press is based on experimental data and theoretical models from [10]).

Table 1 – Details of the irradiation procedure

Incident Ion	Energy		Current, nA	Irradiation time, s	Absorbed dose, MGy	Ion flux, ions/cm^2·s
	MeV/ion	MeV/amu				
H^+	3.00	3.00	100	480	15.15	$1.24\cdot10^{12}$
	1.50	1.50	100	300	14.69	$1.24\cdot10^{12}$
	0.75	0.75	100	60	4.16	$1.24\cdot10^{12}$
He^+	3.02	0.76	120	80	23.12	$1.48\cdot10^{12}$
	2.05	0.51	100	60	15.54	$1.24\cdot10^{12}$
	0.99	0.25	100	60	14.96	$1.24\cdot10^{12}$
C^+	3.00	0.25	30	60	13.64	$0.37\cdot10^{12}$
N^+	3.04	0.22	30	60	16.26	$0.37\cdot10^{12}$
O^+	3.01	0.19	30	60	16.55	$0.37\cdot10^{12}$

3 Results and discussion

Figure1 shows the depth profiles of measured LL intensity and calculated energy deposition for 0.75, 1.5 and 3 MeV protons. The shape of these profiles is comparable with the Bragg's curve. Also, the position of the peaks at the end of the LL intensity profiles (7, 19 and 64 µm for 0.75, 1.5 and 3 MeV protons, respectively) are in rather good agreement with the Bragg's peak positions in the calculated energy loss profiles (6.7, 20.3 and 69.5 µm for 0.75, 1.5 and 3 MeV protons, respectively). However, the Bragg's peak at the end of calculated (dE/dx) profile is very sharp and narrows compared with the corresponding peak in measured profile. Such differences were reported previously [1, 6] and discussed in terms of the overlapping of tracks at high doses.

In the case of low dose irradiated LiF samples, when the overlapping effect between tracks is negligible, a good agreement is observed comparing the calculated and measured radiation damage profiles [1]. When the ion dose increases, the measured profiles become more and more flat because the overlapping of tracks is larger at the Bragg's peak than at the surface of the crystal because difference in the track radius. For high doses (such as curve (c) on figure1), the measured LL intensity profiles become completely flat, and at this stage the overlapping of the tracks in whole penetration depth of the ions leads to a homogeneous and saturated volume.

In figure2 are shown the depth profiles of measured LL intensity and calculated energy deposition for 1, 2 and 3MeV He^+ ions. These ions are expected to deposit almost the maximum energies near the surface region because the incident energy is slightly beyond the energy where the electronic energy deposition reaches the maximum. However, the depth profiles of measured LL intensity and calculated energy deposition have practically the same Bragg's peak positions (1.2, 3.8 and 7.0 µm for 1,2 and 3MeV He^+ ions, respectively). It indicates that the LL method can be used for determining the defect distribution in solids irradiated with low-energy ions (E<MeV/ amu).

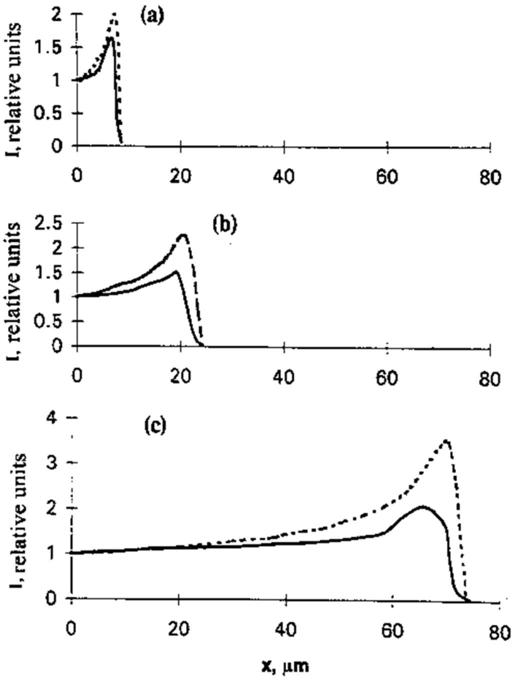

Figure 1 – Calculated energy deposition (interputted line) and lyoluminescense change dependence on thickness of the single crystal dissolved layer (solid line) of LiF bombarded with 0.75 (a), 1.5 (b) and 3.0 (c) MeV protons

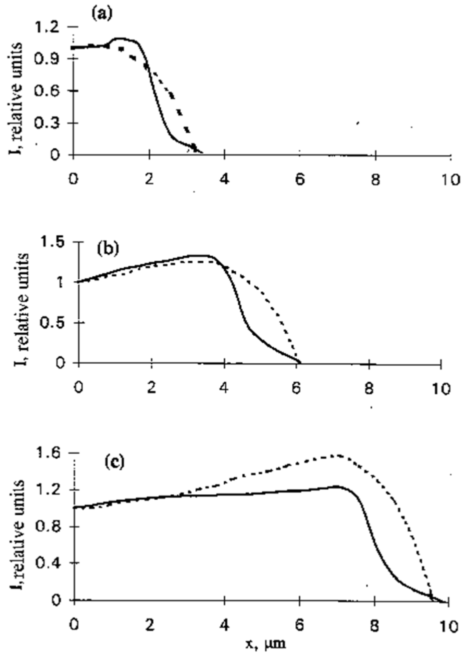

Figure 2 – Calculated energy deposition (interputted line) and lyoluminescence change dependence on thickness of the single crystal dissolved layer (solid line) of LiF bombarded with 1.0 (a), 2.0 (b) and 3.0 (c) MeV He$^+$ ions

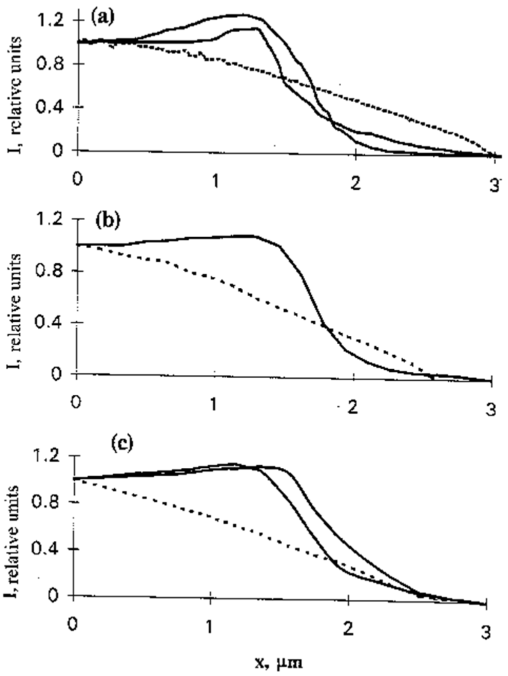

Figure 3 – Calculated energy deposition (interputted line) and lyoluminescence change dependence on thickness of the single crystal dissolved layer (solid line) of LiF bombarded with 3.0 MeV C$^+$ (a), N$^+$ (b) and O$^+$ (c) ions

There are no Bragg's peaks on energy deposition profiles calculated for carbon, nitrogen and oxygen ions (see figure3) because their energy is below the value where the electronic energy deposition reaches the maximum. In spite of this, measured LL intensity profiles exhibit maxims in 1.3 μm region due to the heterogeneous solvent – solid substance interactions during the dissolution of irradiated sample or surface effects on radiation damage. The second of both mentioned possibilities was pointed out in [11] discussing the influence of single crystal surface upon formation of radiation in irradiated alkali halides as well in inorganic salts [12-22]. Direct micro spectrophotometric measurements of radiation damage profiles of accelerated ions LiF [7] also indicates increased concentration of F and F_3 centres in the surface region.

Measured and calculated ranges are presented in table 2. The faint luminescence is observed in the region 0.1 to 0.5 μm (or 0.7 to 13%) beyond the calculated range. This extra luminescence cannot be explained by the small range straggling of about 0.7% and inaccuracy of measurements (approximately 1%). On the other hand, micro spectrophotometric measurements of radiation damage profiles of 180 MeV $Cu^{11+}$and 100 MeV C^{5+} ions in LiF [7] indicated additional coloration caused by F, F_2 and F_3 centres of 43% and 53% deeper than calculated range due to secondary electrons generated by incident ions and recoil atoms. Rather small deviations from calculated ranges in our study are caused by relatively small atomic masses and charges of employed monoenergetic ions.

Table 2 – Calculated and measures ranges

Incident Ion	Calculated range, μm	Measured range, μm	Range deviation	
			μm	%
3.0 MeV proton	72.0	72.5	0.5	0.7
1.5 MeV proton	23.2	23.7	0.5	2
0.75 MeV proton	8.2	8.4	0.2	2
3 MeV He^+	9.5	9.7	0.2	2
2 MeV He^+	6.0	6.2	0.2	3
1 MeV He^+	3.1	3.3	0.2	6
3 MeV C^+	3.0	3.1	0.1	3
3 MeV N^+	2.6	3.0	0.4	13
3 MeV O^+	2.5	2.9	0.4	13

4 Conclusions

We infer that the essential advantage of the LL method, distinguishing it from others (for example, micro spectrophotometric) methods is its high-resolution power. Under gradual dissolution of rotating disk layers' parallel to its surface, monitored by lyoluminescence registration, one can achieve the continuous measurement of defect distribution in a solid with resolving power at a depth of 1 μm. Such resolving power is better than that of any known methods and provides good agreement of measured energy loss profiles with the calculated ones. The defect profiles obtained by means of lyoluminescent method allow to study the defect creation along the whole path of the incident ions and give rather good image of the electronic stepping power. Lyoluminescence measurements seem to be helpful aid for measurement of radiation defects distribution in collision-cascade volume produced by low-energy ions interaction with alkali halides and surface effects on radiation damage.

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