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A novel Al_2O_3 fluorescent nuclear track detector for heavy charged particles and neutrons

G.M. Akselrod [a], M.S. Akselrod [a,*], E.R. Benton [b], N. Yasuda [c]

[a] *Landauer, Inc., Stillwater Crystal Growth Division, 723½ Eastgate Rd., Stillwater, OK 74074, USA*
[b] *Eril Research, Inc., 1110 S. Innovation Way, Ste. 100, Stillwater, OK 74074, USA*
[c] *National Institute of Radiological Sciences, 4-9-1 Anagawa, Inage-ku, Chiba 263-8555, Japan*

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Abstract

A novel Al_2O_3 fluorescent nuclear track detector (FNTD), recently developed by Landauer, Inc., has demonstrated sensitivity and functionality superior to that of existing nuclear track detectors. The FNTD is based on single crystals of aluminum oxide doped with carbon and magnesium, and having aggregate oxygen vacancy defects (Al_2O_3:C,Mg). Radiation-induced color centers in the new material have an absorption band at 620 nm and produce fluorescence at 750 nm with a high quantum yield and a short, 75 ± 5 ns, fluorescence lifetime. Non-destructive readout of the detector is performed using a confocal fluorescence microscope. Scanning of the three-dimensional spatial distribution of fluorescence intensity along the track of a heavy charged particle (HCP) permits reconstruction of particle trajectories through the crystal and the LET can be determined as a function of distance along the trajectory based on the fluorescence intensity. Major advantages of Al_2O_3:C,Mg FNTD over conventionally processed CR-39 plastic nuclear track detector include superior spatial resolution, a wider range of LET sensitivity, no need for post-irradiation chemical processing of the detector and the capability to anneal and reuse the detector. Preliminary experiments have demonstrated that the material possesses a low-LET threshold of <1 keV/μm, does not saturate at LET in water as high as 1800 keV/μm, and is capable of irradiation to fluences in excess of 10^6 cm^{-2} without saturation (track overlap).

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1. Introduction

The radiation dosimetry community has long sought a dosimeter that overcomes the numerous limitations of current passive detector technology. Such a passive integrating detector would be sensitive to charged particles over a broad range of LET, require little or no post-exposure chemical processing, be capable of non-destructive (i.e. multiple) readouts using fully automated equipment, and possess the capability of being erased and reused. Thermoluminescent detectors (TLD) [1] and optically stimulated luminescence detectors (OSLD) [2], while fully reusable and highly sensitive to low-LET radiation, can only measure high-LET radiation of heavy charged particles (HCP) with reduced efficiency and possess little or no sensitivity to neutrons. In addition, TLD can only be read out a single time. Solid state detectors such as CR-39 plastic nuclear track detector (PNTD) [3] possess a sensitivity to radiation with LET in water ($LET_\infty H_2O$) above 5 keV/μm and to neutrons (via neutron-induced proton recoil tracks) [4], but lack sensitivity to lower LET radiation. CR-39 PNTD can only be used once and must be chemically etched prior

* Corresponding author. Tel.: +1 405 3775161; fax: +1 405 7432966.
E-mail address: makselrod@landauerinc.com (M.S. Akselrod).

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to readout. Furthermore, automatic readout of PNTD exposed to isotropic and/or mixed radiation fields, such as those encountered during space flight, is highly problematic. To overcome some of these limitations, a combination of CR-39 PNTD and TLD/OSLD has often been used in the past for dosimetry aboard spacecraft [5] and for accelerator-based radiobiology experiments.

Landauer, Inc. has recently developed a novel fluorescent crystal Al_2O_3:C,Mg [6,7] and demonstrated its applications in volumetric optical data storage [8] and in dosimetry of γ/β radiation [9], neutrons and α-particles [10]. This novel detector material, together with the optical readout technique being developed for its analysis, possesses a number of unique properties that will potentially overcome many of the current limitations of other passive radiation detectors used for space radiation dosimetry and in radiobiology experiments at heavy ion accelerators, while still retaining the advantages of small size, no power, etc. that make passive detectors so useful. The new material, shown in Fig. 1, takes the form of single crystals of Al_2O_3 (aluminum oxide or sapphire) doped with minute concentrations of C and Mg and having a high concentration of aggregate oxygen vacancy defects. While radiation sensitive Al_2O_3:C has been successfully used as a TL [11] and OSL detector [12], the newly developed Al_2O_3:C,Mg fluorescent nuclear track detector (FNTD) has more in common with CR-39 PNTD in that individual particle tracks can be imaged and measured, and particle trajectories can be traced through the volume of the detector.

The mechanism by which ionizing radiation produces tracks in Al_2O_3:C,Mg and the method used to analyze Al_2O_3:C,Mg following irradiation are quite different from those exploited by TLD, OSL or CR-39 PNTD. The presence of oxygen vacancies and Mg ions in the crystal lattice of the Al_2O_3 stimulates production of the new color centers formed by aggregate defects. As with all crystalline materials, the passage of a charged particle through the Al_2O_3 leads to ionization of the crystal and production of free electrons in the conduction band and holes in the valance band. In Al_2O_3:C,Mg, these electrons become trapped in the aggregate oxygen vacancy defects, producing radiochromic transformation of fluorescent color centers existing in the crystal prior to irradiation and created during crystal growth. Later, during analysis of the detector (using a confocal fluorescence laser scanning microscope system), the passage of a focused laser beam over the color centers causes the color centers to fluoresce. The fluorescence is then detected and amplified. Fluorescence is increased in crystal microvolumes where the penetrating particle produces ionization. Analysis (or imaging) of the FNTD consists of scanning a given plane within the crystal volume with a focused laser beam in a raster pattern and measuring the intensity of fluorescence as a function of 3-D location within the crystal. Use of a confocal optical imaging system [13] allows one to detect the laser induced fluorescence from only the focal spot of the laser beam and to spatially discriminate it from the fluorescence induced in the rest of the crystal volume.

Fig. 1. Al_2O_3:C,Mg single crystals and polished detectors.

2. Experimental

2.1. New fluorescent aluminum oxide crystals

New fluorescent aluminum oxide crystals shown in Fig. 1 were developed by doping the material with magnesium and carbon impurities and by growing the crystals in a highly reducing atmosphere [6]. Al_2O_3 is a wide gap insulator ($E_g = 9.5$ eV) and can be engineered to have deep and thermally stable electron and hole trapping centers. Extensive spectroscopic research allowed us to prove that the green–yellow coloration of the "as-grown" crystals is due to aggregate defects formed by association and charge compensation of two oxygen vacancies with two Mg^{2+}-ions substituting Al^{3+}-ions, which are denoted as F_2^{2+}(2Mg)-centers [7].

The color centers in Al_2O_3:C,Mg crystals possess unique electronic and non-linear optical properties. The crystals undergo a bi-stable photo-chromic transformation under blue laser stimulation (reversible changes in optical absorption bands) in conditions of so-called sequential (or resonant) two-photon absorption. Due to their unique optical properties, these crystals were recently suggested as a bitwise volumetric optical medium for next-generation, terabyte capacity data storage [8].

Radiation-induced ionization also causes radiochromic transformations in these crystals [9]. Free electrons and holes generated by ionizing radiation are captured by traps and color centers available in high concentration. Both types of color centers, originally existing in the crystal