

## Development of string-shaped radiophotoluminescence dosimeter for high-radiation field



Fuminobu Sato<sup>a,\*</sup>, Taichi Hashimoto<sup>a</sup>, Shingo Tamaki<sup>a,b</sup>, Sachie Kusaka<sup>a</sup>, Hiroyuki Miyamaru<sup>c</sup>, Isao Murata<sup>a</sup>

<sup>a</sup> Graduate School of Engineering, Osaka University, 2-1 Yamadaoka, Suita, Osaka, 565-0871, Japan

<sup>b</sup> Research Fellow of Japan Society for the Promotion of Science, 5-3-1 Kojimachi, Chiyoda-ku, Tokyo, 102-0083, Japan

<sup>c</sup> Radiation Research Center, Osaka Prefecture University, 1-2 Nakaku, Gakuen-cho, Sakai, Osaka, 599-8570, Japan

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### ABSTRACT

We developed a long and bendable string-shaped dosimeter, 2 mm in diameter. This string-shaped dosimeter, which was made from a mixture of silver-activated metaphosphate glass particles and polylactic acid resin, was fabricated with a filament extruder. The maximum ratio of glass to resin by weight was 20%. The photoluminescence spectra had absorbed-dose-dependent peaks at 635 nm. At doses above 50 Gy, orange photoluminescence could be observed with the naked eye. A one-dimensional (1-D) readout system for string-shaped dosimeters comprised an ultraviolet (UV)-LED, a photodiode, and a drawing machine. In a preliminary experiment, a string-shaped dosimeter was uniformly exposed to <sup>60</sup>Co gamma rays. The position dependency of the detection efficiency of the string-shaped dosimeter was investigated, and its coefficient of deviation was approximately 4%. The string-shaped dosimeter was partly exposed to X-ray beams. The 1-D dose profile of the string-shaped dosimeter was successfully obtained by the readout system. The radiophotoluminescence response was sufficiently related with the absorbed dose in a dose range up to 500 Gy. The spatial resolving power (FWHM) was approximately 3 mm, owing to the width of the slit for the UV illuminator in the readout system. These results indicated that the string-shaped dosimeter was useful in 1-D dose measurement of the high-radiation field.

### 1. Introduction

A radiophotoluminescence (RPL) glass dosimeter is a high-sensitivity, accumulated-type detector (Piesch et al., 1986; Ranogajec-Komor et al., 2008). It has been used as an individual dosimeter in practical applications (Assenmacher et al., 2017). An RPL glass dosimeter is made of silver-activated phosphate glass, in which stable-state photoluminescent centers are formed for exposure to ionizing radiation. Orange luminescence can be observed according to the absorbed dose under ultraviolet (UV) excitation. Silver-activated phosphate glass exhibits superior fading characteristics and fluorescence efficiency when compared with other dosimeter materials. The RPL intensity does not decrease, even under repeated UV excitation. The RPL response exhibits a high sensitivity and linearity over a wide dose range. For this reason, an RPL glass dosimeter can repeatedly read a radiation dose. This property is expected to be useful to various applications of dose measurements (Kurobori et al., 2015).

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(1F) is important at the primary decommissioning stage for the 1F. The development of a new radiation detection system needs for dosimetry in a complex high-radiation field (Ueno et al., 2016). In advanced radiation therapy, new technologies to measure detailed spatial dose distributions for patients are required to assess the therapeutic effect with increased precision (Souza et al., 2017; Martínez et al., 2017; Watanabe et al., 2013). Our research group proposed several visualization technologies based on the RPL characteristics of silver-activated phosphate glass, which can potentially be used for the measurement of high-dose fields within decommissioning of nuclear plants and advanced radiotherapies. The RPL photographing technique that we proposed was applied to the measurement of the spatial dose distribution of high-radiation fields. The fluorescence image of a number of ball-shaped RPL dosimeters was obtained using the RPL photographing technique (Sato et al., 2014). The spatial dose distribution was estimated from the brightness of the obtained fluorescence image (Sato et al., 2015; Zushi et al., 2016).

The fiber-shaped RPL glass dosimeter that we developed was

\* Corresponding author.

E-mail address: [fsato@see.eng.osaka-u.ac.jp](mailto:fsato@see.eng.osaka-u.ac.jp) (F. Sato).

conveniently used for the one-dimensional (1-D) measurement of depth-directional radioactivity distributions (Zushi et al., 2013). In the actual measurement of the depth-directional distribution of radioactive cesium in soil in Fukushima, the fiber-shaped glass dosimeter with a length of 60 mm was embedded into the ground. Measurements of radiocesium-contaminated soil were performed by the fiber-shaped RPL glass dosimeter and a laser microscope. However, the fiber-shaped RPL glass dosimeter was very fragile and needed to be handled carefully; if an attempt was made to bend it, it easily broke. Therefore, we developed a large-length bendable dosimeter. In this paper, we describe the fabrication of a string-shaped dosimeter and this property for dose measurement. Long and bendable string-shaped dosimeters were fabricated with a filament extruder. The 1-D dose measurement was successfully performed with a string-shaped dosimeter and a readout system.

## 2. Methods

### 2.1. Materials

The string-shaped dosimeter was made from a mixture of RPL glass particles and polylactic acid (PLA) resin powder. The details of the production method of an RPL glass rod have been described in previous papers (Lee et al., 2011). The rod of RPL glass was made from reagent-grade powders using a melting method.  $\text{NaPO}_3$  (100 g),  $\text{Al}(\text{PO}_3)_3$  (104 g), and  $\text{AgCl}$  (4.4 g) were added to an alumina crucible. The alumina crucible was placed in an electrical furnace, and subsequently, its temperature was gradually increased to 1200 °C over the course of 10 h. The melting glass was maintained at this temperature for 5 h to facilitate homogenization. After homogenization, the melted mixture was slowly cooled to room temperature over the course of 10 h. The cooled glass was cut into pieces with a rotating diamond saw blade. The glass pieces were pulverized by a mortar, and the pulverized particles were classified using 75 and 150  $\mu\text{m}$  sieves.

The characteristics of the RPL glass particles were reported previously (Sato et al., 2014). Silver atoms in silver-activated phosphate glass exist in a stable state as  $\text{Ag}^+$ . If ionizing radiation irradiates the glass, electrons and positive holes are generated. The electrons are trapped by  $\text{Ag}^+$ , which is converted into metastable  $\text{Ag}^0$ . Furthermore, after the positive holes are trapped by  $\text{PO}_4$  tetrahedra, as time passes, positive charge is transferred to  $\text{Ag}^+$ , which becomes metastable state  $\text{Ag}^{++}$  (Knežević et al., 2013). These  $\text{Ag}^0$  and  $\text{Ag}^{++}$  states both exhibit orange fluorescence characteristics under UV excitation. The fluorescence efficiency is proportionally related to the absorbed radiation dose. The resulting RPL center does not lose the absorbed dose information. Thus, the information can be read repeatedly. Heat treatment (e.g., 400 °C for 1 h) is required to destroy the RPL center.

PLA pellets (4043D, NatureWorks, LLC, MN, USA) were prepared. PLA is a thermoplastic aliphatic polyester, whose density is 1.24 g/cm<sup>3</sup>. PLA can be processed into strings (for example, using conventional melt spinning processes) and films, like most thermoplastics. The melting point is 210 ± 8 °C. Therefore, PLA is widely used as a material for 3-D printing with thermoplastics. In addition, PLA has a higher affinity for other materials than acrylonitrile butadiene styrene resin, which is one of materials used for 3-D printing. The PLA pellets were pulverized into particles using a crusher (TPH-02, AS ONE Corp., Osaka, Japan) with liquid nitrogen. The PLA particles were classified using 75 and 150  $\mu\text{m}$  sieves.

### 2.2. Fabrication

String-shaped dosimeters were fabricated from a mixture of RPL glass particles and PLA powders using a filament extruder (Filabot EX2 filament extruder, VT, USA). The filament extruder was one of plastic-extruding machines and was used to produce filament of 3-D printers. A nozzle, 2 mm in diameter, was set on the filament extruder. It was

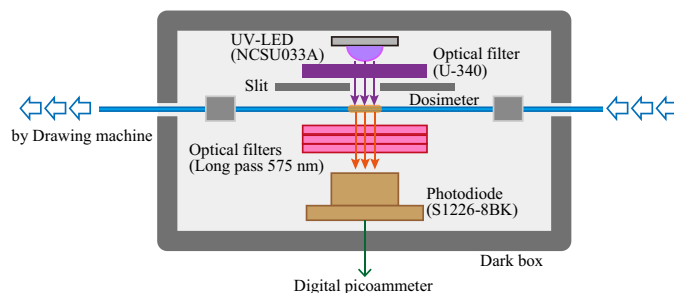


Fig. 1. Layout of a homemade 1-D dose readout system for string-shaped dosimeters. This readout system was composed of a UV-LED, photo sensor, and drawing machine.

found from the process of trial and error that the appropriate temperature for extruding the mixture was 200 °C. The concentration of the RPL glass particles was up to 20% by weight.

### 2.3. RPL readout system

The schematic drawing of a homemade readout system is shown in Fig. 1. The string-shaped dosimeter was set on the sample stage in the dark box, and pulled out by the drawing machine with a stepper motor (SGSP-60YAW-OB, Sigma Koki Corp., LTD., Saitama, Japan), at a drawing speed of 0.3 mm/s. An LED (NCSU033A, Nichia Corp., Tokushima, Japan) with a wavelength of 365 nm was used for the excitation on the detention part of the string-shaped dosimeter. The temperature of the LED was controlled to maintain the stability of the photon emission. The UV intensity on the sample through a band pass filter (U-340, Hoya Corp., Tokyo, Japan) was approximately 10 mW/cm<sup>2</sup>. A slit, by which the UV illuminating area on the sample was controlled, was placed in the front of the sample stage. The width of the slit was adjusted to be 3 mm. The RPL was detected through optical filters (Long pass filter OD4-575 nm, Edmund Optics Inc., NJ, USA) with a photodiode (S1226-8BK, Hamamatsu Photonics K.K., Shizuoka, Japan). The current of the photodiode was measured with a picoammeter (8240, ADC Corp., Saitama, Japan). The current and the position of the sample were simultaneously recorded in a personal computer at intervals of 1 s.

### 2.4. Irradiation test

We examined the performance of the dose readout of the string-shaped dosimeter, which was uniformly exposed to gamma rays with doses of up to 500 Gy. The 50 cm-string-shaped dosimeter, which was ring-formed, was annularly placed in the center of an intense <sup>60</sup>Co gamma ray source.

The maximum of the X-ray tube voltage was 45 kV, and its maximum current was 30 mA. The electron beam irradiated a Cu target, and X-rays propagated through a 125- $\mu\text{m}$ -thick beryllium window and were collimated by slits made of stainless steel. The width of the slit was adjusted to be 10 mm. The absorbed doses at sample positions were controlled by the irradiation time.

## 3. Results and discussion

Fig. 2 shows a photograph of a long and bendable string-shaped dosimeter. The diameter of the string-shaped dosimeter slightly depended on the positions owing to the fabrication process. The average and the coefficient of the deviation of the diameters were 2.0 mm and 3%, respectively. The surface of the string-shaped dosimeter was not smooth. It was found from the surface observation using a scanning electron microscope (JSM-5600LV, JEOL, Tokyo, Japan) that some RPL glass particles were protruding from the surface. The density of the string-shaped dosimeter with 20 w% RPL glass was estimated to be approximately 1.5 g/cm<sup>3</sup>. Fig. 3 shows photographs of the RPL in the

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