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Development of radiophotoluminescence glass dosimeter usable in high temperature environment

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HIGHLIGHTS

- A new radiophotoluminescence (RPL) glass dosimeter was developed for use in high temperature conditions.
- The RPL intensity of the glass dosimeter was satisfactorily sustained at 573 K for 3 h.
- The RPL response of the glass dosimeter had satisfactory linearity in the dose range from 10 to 10^4 mGy.

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ABSTRACT

A new radiophotoluminescence (RPL) glass dosimeter was developed for use in high temperature conditions such as nuclear emergencies. Its glass material was successfully made by a melting method from reagent grade powder of $Ca(H_2PO_4)_2$, $NaPO_3$ and AgCl. The new RPL glass dosimeter expectedly emitted orange photons for exposure to UV light after gamma-ray irradiation. It was confirmed that its RPL intensity was proportional to absorbed dose in the range from 10 to 10^4 mGy. As for its temperature-proof performance, it was found that the RPL sensitivity hardly changed at 573 K for 3 h but gradually went down 25% for 50 h.

1. Introduction

In the nuclear disaster at the Fukushima Daiichi nuclear power plant, a large amount of radioactive substance was released into the environment. The high-level radioactivity seriously prevents engineers from approaching the broken nuclear reactors. In addition, although precise dose measurement is required for radiation safety management, the high temperature and humidity conditions make it difficult to use electronic radiation instruments.

Thermoluminescence dosimeters (TLDs) have the advantages of high sensitivity, wide measurable dose range and others. However, some TLDs have peaks above 373 K in their glow curve (Datz et al., 2010), and they are not suitable for use in high temperature environment because of the large fading of the main dosimetry peak at high temperature. Development of a new dosimeter usable at high temperature is one of many tasks for decommissioning the broken nuclear power plant.

Fundamental characteristics of a radiophotoluminescence (RPL) glass dosimeter were investigated for personal dosimetry and environmental monitoring (Piesch et al., 1986). The RPL glass dosimeter has excellent characteristics such as small fading effect at room temperature (Lee et al., 2009), good dose linearity and high reproducibility (Ranogajec-Komor et al., 2008). A commercially available RPL glass dosimeter is made of a silver activated metaphosphate glass, of which composition is $xNaPO_3 \cdot (1-x)\,Al(PO_3)_3$ ($0 \le x \le 1$, Na–Al glass) from the viewpoint of the RPL efficiency and effective atomic number (Miyamoto et al., 2011). However, the Na–Al glass dosimeter is difficult to use at high temperature because of the fading effect over 423 K.

Phosphate glass in general is formed by anions that are made of PO_4 tetrahedra and linked metal cations. Large concentration of alkali metal such as Li^+ and Na^+ is allowably incorporated in the phosphate glass. In a silver activated phosphate glass, silver atoms exist uniformly and stably in the form of Ag^+. Yokota and Imagawa

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elucidated that electrons and holes were caused by ionizing radiation and diffused in the phosphate glass, and induced the formation of Ag^0 and Ag^{++} as RPL centers (Yokota and Imagawa, 1967). The RPL centers have high stability at room temperature and emit orange photons for exposure to UV light. The holes trapped in Ag^{++} centers are believed to be transferred from neighboring PO_4 tetrahedra. The reaction speed at the room temperature is slow owing to low mobility of Ag^+ and hole in the phosphate glass. In actual use, therefore, a process of preheating at 343–373 K accelerates the formation of the Ag^{++} centers. This charge transfer process is the main cause of the known RPL buildup. Meanwhile, the reaction speed for the formation of Ag^0 center is faster than that for Ag^{++}. The Ag^0 center is easily formed at room temperature. Moreover, Dmitryuk et al. have proposed that the RPL center is originated from Ag_3^{++} formed by the combination of Ag^0 and double Ag^+. The mobility of Ag^+ is one of important parameters of RPL center formation (Dmitryuk et al., 1996). In the further heating of annealing process, accumulated RPL centers disappear through the recovery of Ag^+ from Ag^{++} or Ag^0. The formation and thermal tolerance of RPL centers are related to the molecular structure of phosphate glass. The characteristics of RPL centers can be partially controlled by the content of the linked metal cations such as alkali metals, alkali earths and aluminum.

We have tried to develop a new glass dosimeter usable in high temperature conditions such as nuclear emergencies. A temperature-proof RPL glass dosimeter was well synthesized and its characteristics were examined in this study.

2. Experimental

Fig. 1 shows mean molar volume of metaphosphate glass (Tsuchida et al., 2011). Ag-doped $NaPO_3$ glass is crystalline material with RPL effect. The mobility of Na^+, Ag^+ and hole increases with the sodium concentration. Therefore, sodium cation plays an important role in the RPL center formation (Dmitryuk et al., 1996). However, only $NaPO_3$ cannot be used as material for a practical RPL glass dosimeter owing to its deliquescence. The molar volume of $Al(PO_3)_3$ is fairly large owing to the conformation of Al^{3+} with triple bonding. In fact, a commercially available glass dosimeter is made from $NaPO_3$ and $Al(PO_3)_3$ and it has appropriate hardness and water resistance. The RPL centers in the Na–Al glass are stable at room temperature and the fading hardly occurs until 423 K.

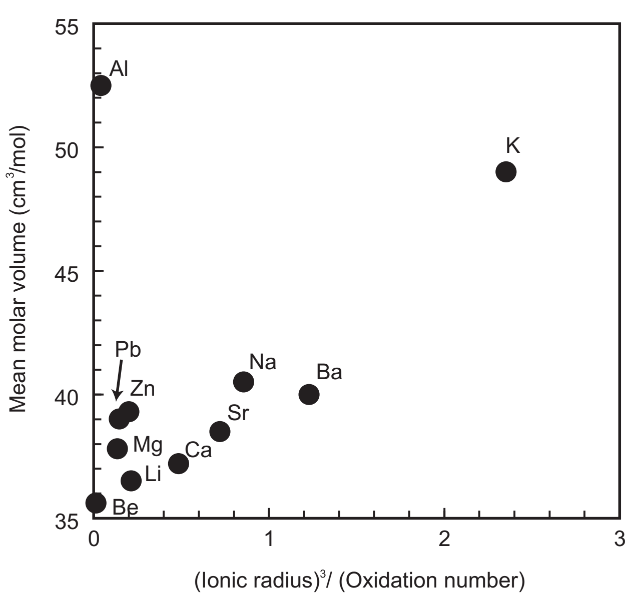

Fig. 1. Mean molar volume of various metaphosphate glass.

From a viewpoint of molar volume of metaphosphate glass, $xNaPO_3 \cdot (1-x)Ca(PO_3)_2$ $(0 \le x \le 1$, Na–Ca glass) is one of candidate materials for RPL glass dosimeters usable in high temperature environment. The molar volume of $Ca(PO_3)_2$ is comparatively smaller than that of $Al(PO_3)_3$. The distance between Ag^+ and neighboring molecules is short and Ag^+ is moderately secured by neighboring molecules. Therefore, the temperature tolerance of the RPL centers might be higher than that of the Na–Al glass. $Be(PO_3)_2$ and $Li(PO_3)$ with small molar volume are also candidate materials for temperature-proof glass dosimeters. However, beryllium compounds are difficult to handle owing to their toxicity. Also, a RPL glass dosimeter containing Li for neutron dosimetry was developed by Maki et al., but its temperature tolerance has not been examined yet (Maki et al., 2011a).

Ag-doped Na–Ca glass was made from reagent grade phosphate powder by a melting method (Lee et al., 2011). The kinds of the reagent grade powders were calcium dihydrogenphosphate $[Ca(H_2PO_4)_2]$, sodium metaphosphate ($NaPO_3$) and silver chloride (AgCl). Approximately, 85 g of $Ca(H_2PO_4)_2 \cdot H_2O$, 10 g of $NaPO_3$ and 0.2 g of AgCl were added to an alumina crucible. The alumina crucible was set in an electrical furnace. In a pretreatment, water was removed from the mixture through the heat-treatment at 523 K for 15 min. The mixture was melted and kept at 1473 K for 10 h in the crucible. After homogenization, the melting mixture was poured into a brass mold preheated at 773 K. The temperature of fabricated glass was kept at 793 K for 24 h and it was slowly cooled down to room temperature in 10 h. The cooled glass was cut into small pieces of $5 \times 5 \times 1$ mm^3 with a rotating diamond saw blade. The surface of the glass plates was polished with alumina and cerium lapping disks. The atomic weight composition in the Na–Ca glass dosimeter sample was as follows: O (49%), Na (3%), P (32%), Ca (16%) and Ag (0.1%).

The Na–Ca glass dosimeter samples were irradiated with ^{60}Co gamma-rays up to 10 Gy. Moreover, the Na–Al glass dosimeters were used as references. The atomic weight composition of the Na–Al glass dosimeter was as follows: O (51%), Na (11%), Al (6%), P (32%) and Ag (0.2%). There was not a large difference in the RPL spectra and the sensitivity among the synthesized Na–Al glass dosimeter and a commercial dosimeter FD-7. After the preheating of the glass dosimeters for the build-up of RPL centers with different temperatures (300–700 K), their responses were examined with a RPL readout system (Maki et al., 2011b). The uncertainty in the RPL measurement was less than 5%. It is known that the RPL has some different decay-time components. The component with a decay time of 2–4 μs is proportional to the concentration of RPL centers, i.e., radiation dose. The wavelength spectra of RPL photons from the glass dosimeter samples were measured with a calibrated photon spectrometer.

3. Results and discussion

Fig. 2 shows an example of typical radiophotoluminescence spectra of Na–Ca glass dosimeter samples to gamma-rays. A pulse laser of 355 nm in wavelength was used as a UV light excitation source. It was found that the RPL spectrum of the gamma-ray-irradiated Na–Ca glass dosimeter sample had a large peak around 650 nm in wavelength. It was also confirmed from comparative similar experiments that there was not a large difference in the shape of the RPL spectrum between the Na–Ca and Na–Al glass dosimeter. For non-irradiated glass, there was intrinsic photoluminescence which was scarcely connected to radiation dose. For Ca–Na glass without Ag, moreover, the intrinsic photoluminescence was hardly observed for the UV light excitation. It is