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Radio-Photoluminescence Glass Dosimeter (RPLGD)

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1. Introduction

Radiation is a type of the energy transport. It produces ionization, scintillation, and luminescence when radiation interacts with matter. By detecting these phenomena from the response of the dosimeter after exposure, one can acquire an understanding on the types and the intensity of the radiation.

Solid state dosimeters can be divided into two categories, active dosimeters and passive dosimeters. When radiation interacts with medium inside the dosimeter, the active dosimeter transfers radiation intensity into the pulse of electric signals. Based on those signals, users can determine the types and the intensity of the radiation. As for passive dosimeters, radiation interaction is detected through certain physical processes after radiation interacts with medium in the dosimeter. From the physical processes, users can also determine the types and the intensity of the radiation. Active dosimeters are used for dose measurements in areas with unknown radiation level to gather the radiation information immediately. Therefore, the proper radiation protection actions can be initialized. The common active dosimeters in the market are gas-filled counters, scintillation counters, and semi-conductor detectors…etc. On the other hands, passive dosimeters are often used as periodic radiation monitor for people work in the radiation environment to monitor the cumulated dose and the types of radiation. They can be used as personal dose measurement, long-term environmental radiation dose monitor…etc. The film badge, Thermoluminescence Dosimeter (TLD), Optically Stimulated Luminescence Dosimeter (OSLD), and Radio-photoluminescence Glass Dosimeter (RPLGD) are commonly used passive dosimeters. In clinics, many different kinds of dosimeters are applied in the procedures to verify dose delivery accuracy, to obtain dose to critical areas or organs, and to verify machine output for QA purposes.

For TLD, OSLD, and RPLGD, when radiation interacts with the medium in the dosimeters, part of the absorbed energy are first stored in a metastable energy state of the medium. Then some of this energy can be recovered later as visible light after proper physical process, such as heating.

2. Radio-Photoluminescence Glass Dosimeter (RPLGD)

OSLD is made of the same luminescent material as one used in TLD. The only differences are different excitation source and different readout technique used. However, RPLGD uses

glass compound as the luminescent material and applies different excitation method along with different readout technique. In 1949, Wely, Schulman, Ginther, and Evans manufactured the first RPLGD system (Yokota). Schulman applied this system in radiation dose measurement in 1951 (Yasuda, Troncalli). The luminescent material used by Schulman was a compound glass of 25% of KPO_3, 25% of Ba $(PO_3)_2$ and 50% of Al $(PO_3)_2$, with proper amount of $AgPO_3$ to form silver activated phosphate glass. It is very difficult to measure dose under 1 mGy with Schulman's RPLGD system, because it has a high pre-dose (residual dose). Pre-dose is the phosphorescence light emitted from RPLGD without any irradiation and excitation process. It is the minimum radiation can be measured with RPLGD. Besides, because of the pre-mature luminescence measurement technique and the poor quality of excitation source for color centers, the measurement accuracy with Schulman's RPLGD is very poor. Therefore, RPLGD is not a popular dosimeter in day to day applications in those days.

However, there are many researchers continue to devote in the developments of RPLGD and its readout system; including people at Asahi Techno Glass Corporation (ATGC) in Japan, at Toshiba Corporation in Japan, and at Karlsruhe Nuclear Research Center (KNRC) in Germany. The developments of new generation RPLGD and readout system were completed in 1990 (Piesch). Table 1 shows the types and compositions of the glass luminescent material developed by ATGC and Toshiba. The excitation source was changed from ultra-violet into pulse ultraviolet laser. The improvements in the glass material and in readout system make the RPLGD capable for lower dose (10 µGy) measurement with excellent accuracy (A. T. G., Corporation Chiyoda Technol).

TLD is still the major dosimeter used for personal dose monitor and for dose verification in diagnostic radiology and in radiotherapy in nowadays. The major problem with TLD is its non-repeatable readout for the measurements. Based on the preliminary report by Hsu et al on the study of the characteristics of RPLGD in radiation measurement, it proves that the radiation detection characteristics of RPLGD are superior to that of TLD (Hsu). Therefore, in the near future, RPGLD will become one of the important dosimeters for dose measurement and radiation detection in the field.

The work on the radiation measurement with self-manufactured RPLGD by Schulman in 1951 opened the history of RPLGD applications in dose measurement (Yasuda, Troncalli). After exposed to radiation, stable color centers are formed in the glass and more color centers are formed with increasing radiation intensity. After irradiated by ultraviolet light, color centers are excited and emit 600 nm to 700 nm visible orange light (Burgkhardt). It is called radio-photoluminescence phenomenon. The amount of orange light emitted from RPLGD is linearly proportional to the radiation received; therefore, it is suitable for long term personal dose monitor or environmental radiation monitor. RPLGD is used in Japan for over 80% of radiation workers as an external dosimeter (Corporation Chiyoda Technol).

3. Principle of RPLGD and its readout methods

The basic principle of RPLGD is that the color centers are formed when the luminescent material inside the glass compound exposed to radiation and fluorescence are emitted from the color centers after irradiated with ultra-violet light. The excited electrons generated from the color centers return to the original color centers after emitting the fluorescence. This process is called radio-photoluminescence phenomena. Because the electrons in the color centers return to the electron traps after emitting the fluorescence, it can be re-readout for a single irradiation.

glass series	composition ratios (mol%)							
	Li	Na	P	O	Al	Ag	Mg	Ba
FD-1	3.7	-	33.4	53.7	4.6	3.7	-	0.9
FD-3	3.6	-	34.5	53.5	5.1	3.3	-	-
FD-4	3.5	-	34.0	52.7	5.0	4.8	-	-
FD-5	-	9.0	33.1	51.3	6.1	0.5	-	-
FD-6	-	6.6	33.2	51.4	5.5	1.4	1.9	-
FD-7	-	11.0	31.5	51.2	6.1	0.2	-	-

Table 1. The types and compositions of silver activated phosphate glass

Figure 1 shows the old RPLGD readout technique (Piesch). The pre-dose M_0 (M_0 (t_0) = I_2 x Δt) was obtained with photomultiplier tube (PMT) first. After RPLGD irradiated by the radiation, the total light intensity is M_1 (M_1 = I_1 x Δt). The "actual" light intensity from the irradiation, M, is M_1 - M_0 = (I_1 x Δt) – (I_2 x Δt). The radiation dose can then be estimated from M. The traditional way to calculate the light intensity is to subtract the pre-dose reading (M_0) from the total reading (M). With the traditional readout technique, if the glass surface is covered with dust or other material the pre-dose reading (M_0) and the total dose reading (M) are both affected and results in a large error for dose estimation. Therefore the old RPLGD readout technique will not measure the dose accurately.

In 1990, a new RPLGD readout system was developed by the cooperation of ATGC (Japan) and KNRC (Germany). The major modification in this new system is to use pulse ultraviolet laser as excitation source, instead of ultra-violet light. The intensity, the excitation time and position of the pulse ultra-violet laser can be accurately controlled. Traditionally, it takes seconds for the unit to count the excitation time; however, it has changed to micro second (μs) for the new system. The readout time is decreased rapidly with the new system. Furthermore, with a collimated laser beam, the laser can be delivered to the exact position in the glass. The radiation energy can also be estimated accurately with the energy compensator filter.

With the pulse ultra-violet laser excitation system, decay curve of fluorescence can be divided into three portions according to the fluorescence decay time of RPLGD. They are (1) pre-dose or the light signal emitted from the impurity covering the glass surface, (2) the light signal from color centers formed by radiation, and (3) the light signal emitted from predose after long time decay.

Any signal detected within the fluorescence decay time between 0 to 1 μs, the readout system mark it as the light signal from pre-dose or from the impurity on the glass surface.

The readout system takes light signal emitted in the fluorescence decay time between 1 to 40 µs as the signal from radiation exposure. For light signal emitted in the fluorescence decay time up to 1 ms, the readout system takes it as the signal produced by pre-dose with long decay time characteristics. The characteristics of the fluorescence decay curve are illustrated in figure 2.
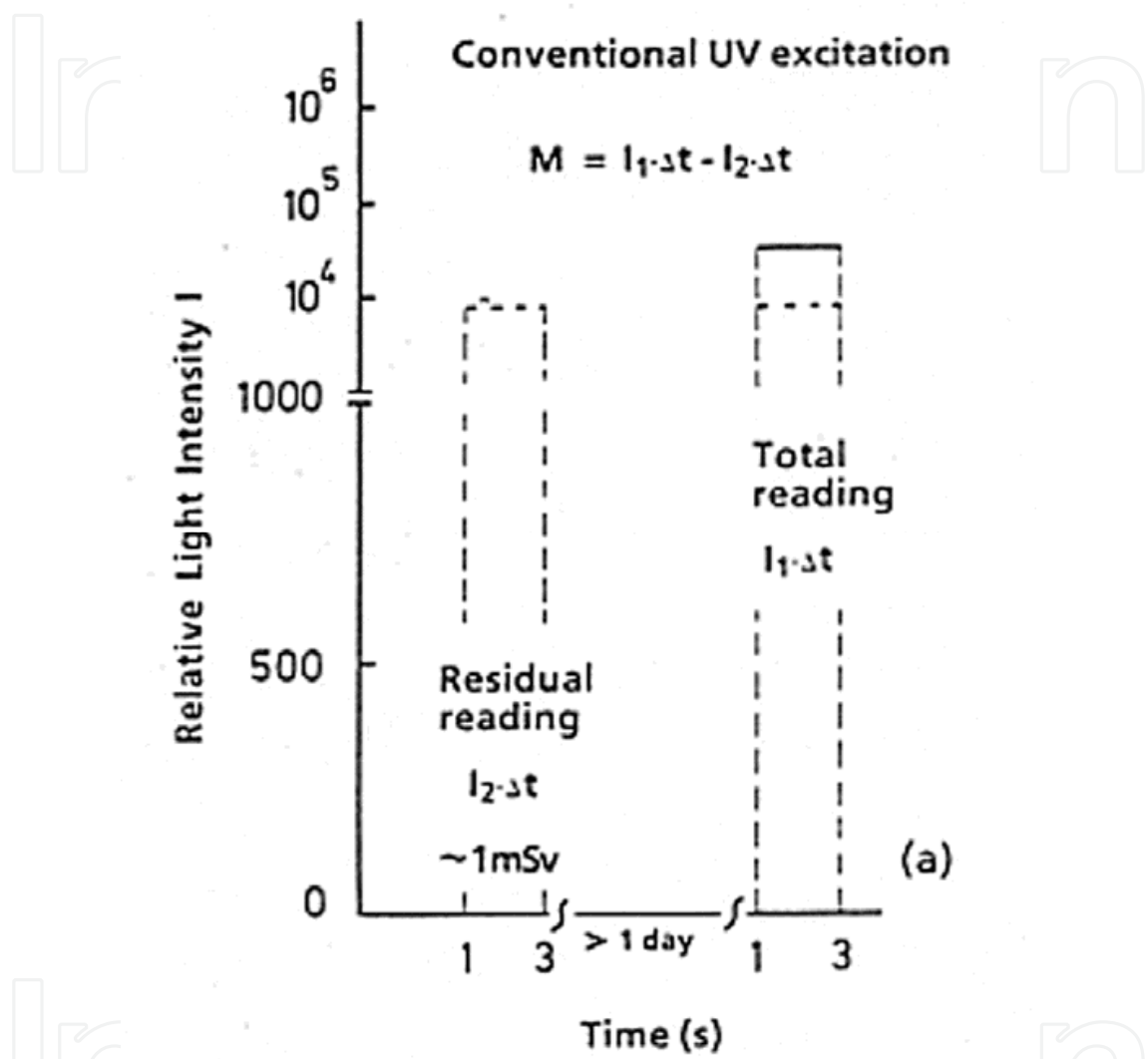

Fig. 1. Old readout technique for RPLGD (Piesch)

In figure 3, the area of F_1 is the integral of fluorescence decay curve between t_1 (1 µs) and t_2 (40 µs) and it is the luminescence signal produced by radiation. However, there are pre-dose signals included in the lower half part of F_1, therefore, one should subtract this portion from F_1 to obtain the "actual" luminescence signal emitted by exposure. The way to subtract the pre-dose signal is to find F_2 from the longer fluorescence decay curve of pre- dose. The area of F_2 is the integral between t_3 and t_4 where time between t_3 and t_4 and t_1 and t_2 is the same, 39 µs. From the proportional relationship of trapezium area, it shows the area of pre-dose in F_1 is $F_2 \times f_{ps}$ (f_{ps} is the conversion factor for trapezium area). Therefore, the actual luminescence signal from the color centers is $F_1 - F_2 \times f_{ps}$. The exposure received by RPLGD can be obtained from the luminescence signal emitted.
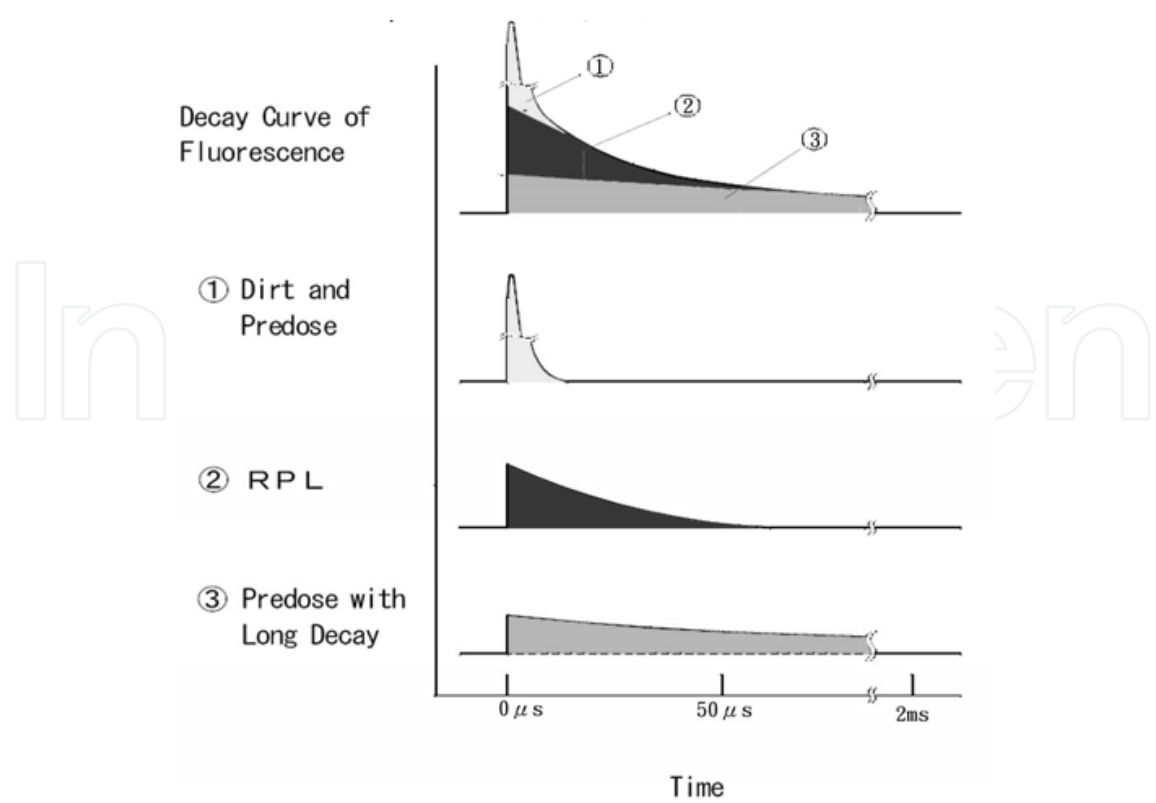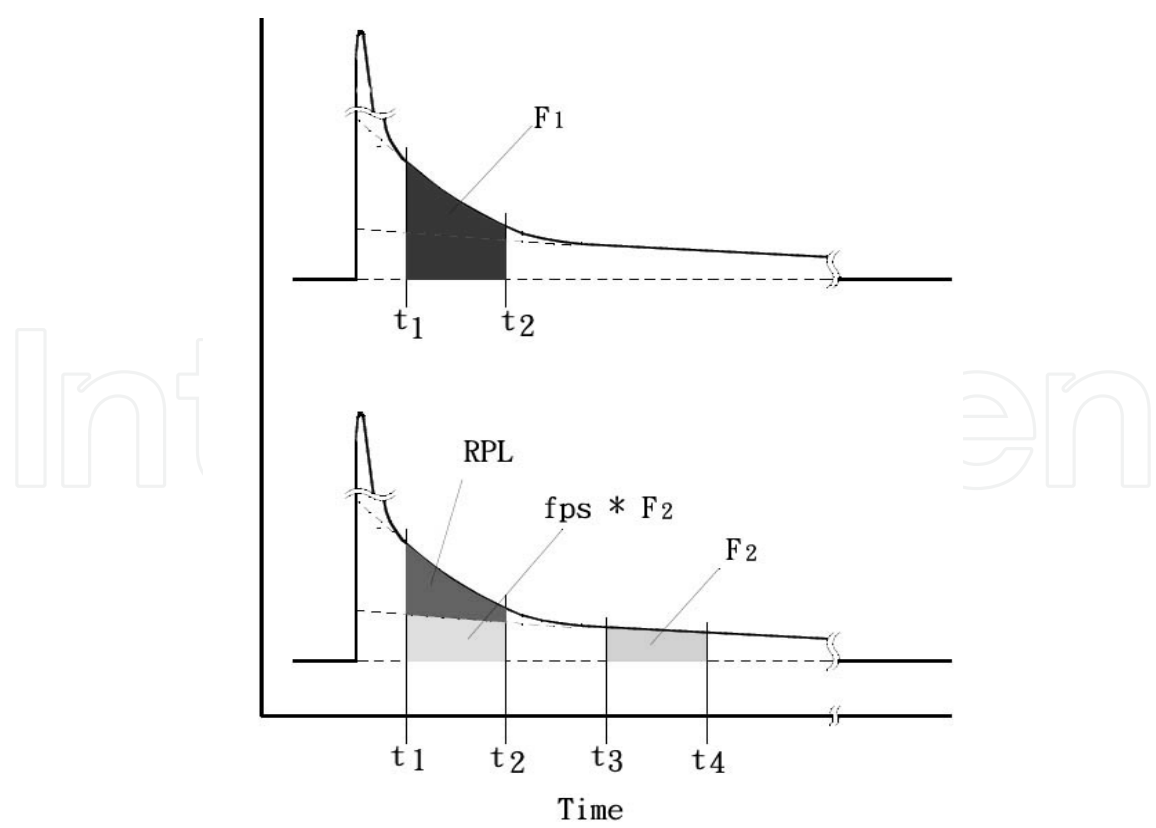
Fig. 2. The luminescence decay curve of RPLGD (A. T. G.)

Fig. 3. The readout technique with pulse ultra-violet laser (A. T. G.).

4. Chemical characteristics of the silver ions

The color centers were structured at the silver activated phosphate glass. The numbers of ionic silver relate to energy levels in color centers and the numbers of electron trap(s). The numbers of electron trap(s) increase with increasing numbers of ionic silvers. However, excessive numbers of ionic silver decrease the penetration efficiency of the pulse ultra-violet laser and increases energy dependence. Therefore, a proper ratio of ionic silver is required for the best luminescence and excitation efficiency (Yokota).

At present, the most common type of glass in RPLGD for radiation dose measurement is FD-7. The $AgPO_4$ in silver activated phosphate glass of FD-7 can be viewed as Ag^+ and PO_4^-. When the tetrahedron of PO_4^- is exposed to the radiation, it loses one electron and forms a "positron hole". The electron released from the PO_4^- will combine with Ag^+ to form an Ag^0. Similarly, hPO_4 ("hole" formed after PO_4^- loses one electron) will combine with Ag^+, and then gains a "positron hole" to become an Ag^{2+}. Both Ag^0 and Ag^{2+} can form color centers as shown in Figure 4.

$$Ag^+ + e^- \longrightarrow Ag^o \ (\text{electron trap})$$
$$Ag^+ + hPO_4 \longrightarrow PO_4 + Ag^{2+} \ (\text{hole trap})$$
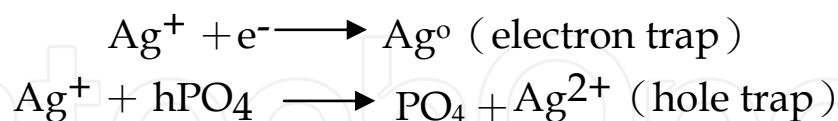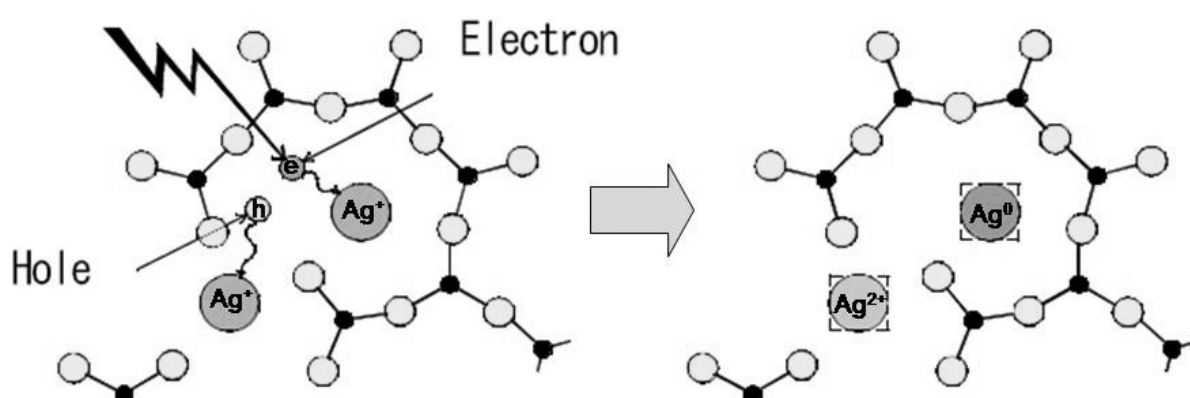
Fig. 4. The color centers formation mechanism of FD-7 (A. T. G.)

After exposure, the Ag^+ at valence band of silver activated phosphate glass combines with electron released from both PO_4^- and hPO_4 (formed by PO_4^-) to become color centers (Ag^0 and Ag^{2+}). When these color centers excited by 337.1 nm pulse ultra-violet laser, the electrons in Ag^0 and Ag^{2+} excited to higher energy levels and emit 600 – 700 nm visible orange light, then return to the original color centers. Energy gained by electrons from the pulse ultra-violet laser is not high enough to let electron escape from color centers. Therefore these electrons will not return to the valence band of the glass material directly. For electrons to gain enough energy to return to the valence band, we need to anneal RPLGD at 400^0C for one hour. The color centers won't disappear after readout; hence, RPLGD can be read repeatedly. Figure 5 shows the energy levels of RPLGD.
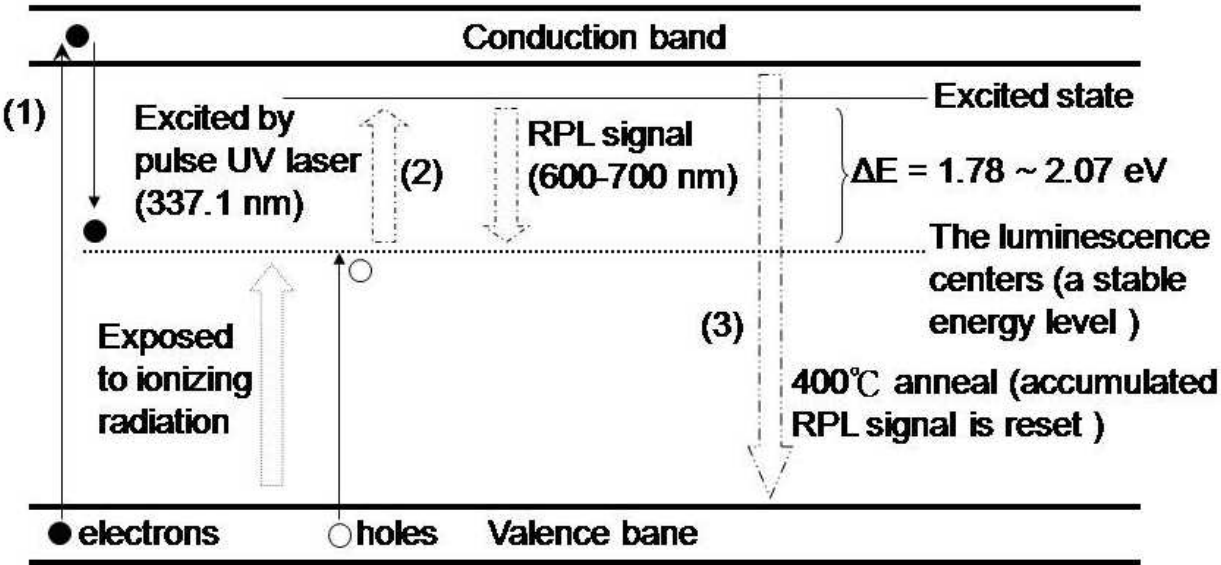
Fig. 5. The energy levels of RPLGD. (1) After RPLGD being exposed, Ag^+ at the valence band combines with electron released from PO_4^- and hPO_4 formed by PO_4^- to become a color center. (2) After electron at color center excited by 337.1 nm pulse ultra-violet laser, it will be excited and emits 600 – 700 nm visible orange light, then return to the original color centers (3)After annealing at 400^0C for one hour, the electron at color centers returns to the valence band of luminescence material (Hsu).

5. The radio-photoluminescence model

The luminescence materials used in either TLD or OSLD have an ordered crystal structure with lattice defects. From the glow curve, which is generated after annealing, one has the information on the electron distribution functions at different energy trap(s). The luminescence models for TLD and OSLD are developed based on this information. However, RPLGD is a mixture of inorganic amorphous solid and does not have lattice structure and lattice luminescence centers. Therefore we cannot get the information on electron trip(s) distribution function to establish the luminescence model for RPLGD. We can only establish the radio-photoluminescence model based on the energy of the excitation source and the energy of the released visible light.

After excited with 337.1 nm pulse ultra-violet laser, RPLGD emits 600 – 700 nm visible lights. From the emitted lights we know the energy gap between the excited energy levels which electrons jump to and the energy levels at color centers is between 1.78 and 2.07 eV. Becker assumed there are many continuous energy levels at the color centers of the RPLGD (Becker), as shown in Figure 6. It shows the electrons in the valence band are excited to the conduction band after irradiation. When electrons return to the valence band, portions of electrons are captured by the electron trap(s) located at P shell and Q shell, and then form color centers. After excitation, the electrons in color centers jump to higher energy level, emit fluoresce, then return to the original color centers. RPLGD is manufactured via the process of melting various compounds under high temperature, different from the manufacture process of TLD or OSLD which is via process of long-crystal formation. Hence, the color centers of PRLGD are not built at the lattice. There are no formal reports on the

luminescence model for RPLGD. We believe that the color centers of RPLGD may be structured among the orbital electrons in the compound. The various continuous energy levels are formed with different bonding structures among elements. Those energy levels can store free electron energy which is produced by the excitation process. Therefore, its excitation energy gap has a continuous value (from 1.78 to 2.07 eV) which releases 600 nm – 700 nm visible lights.
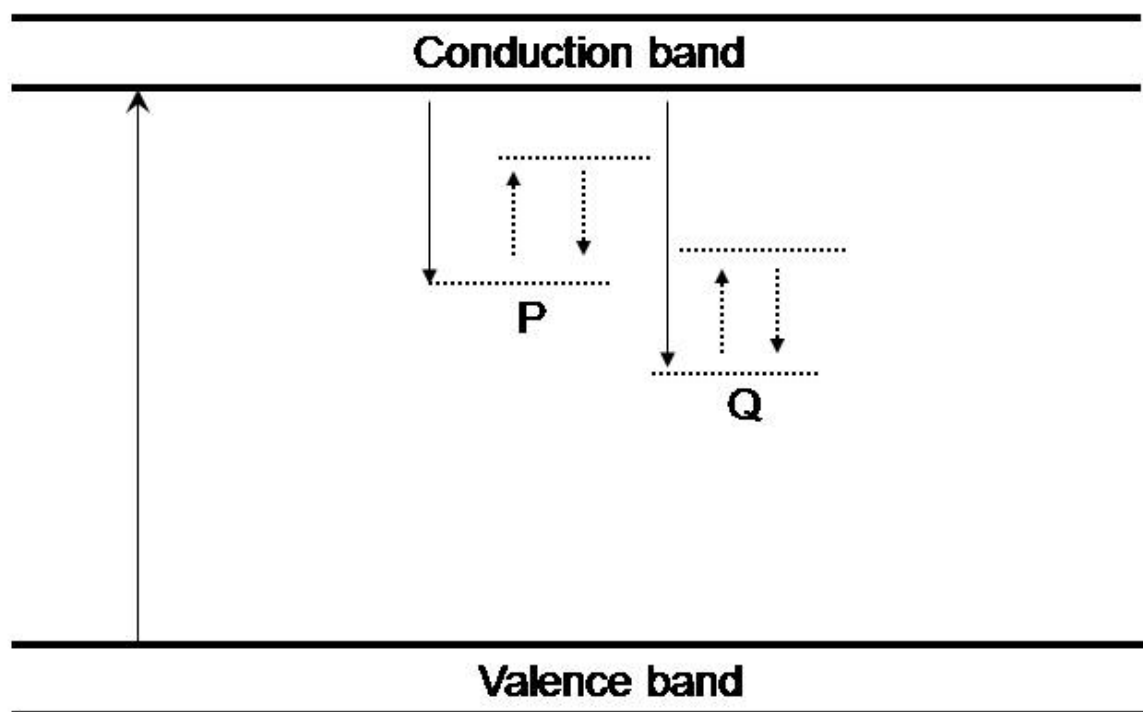

Fig. 6. There are many continuous energy levels in RPLGD color centers.

6. Physical characteristics of radio-photoluminescence glass dosimeter

The pulse ultra-violet laser excitation system improves the readout accuracy of RPLGD and also shortens the readout time. The improvement in the luminescent material lowers the detectable dose limit. These improvements make the applications of RPLGD in radiation measurements growing rapidly. There are three major types of RPLGD in the market; the SC-1 for environmental radiation dose monitor; the GD-450 for personal external radiation dose monitor; and the Dose Ace for research purposes. All those three types use FD-7 glass, manufactured by Asahi, Japan, as shown in Figure 7.

The SC-1 is a plate-type RPLGD with outside capsule volume of 30 x 40 x 9 mm³.The dimension of FD-7 glass inside the capsule is 16 x 16 x 1.5 mm³. There are two layers of tin filters, one on the top and another at the bottom, over of the capsule with a dimension of 0.75 mm and 3 mm respectively. These tin filters are used as energy compensator to estimate the radiation energy and to lower the energy dependence effect. The FD-7 in GD-450 has a dimension of 33 x 7 x 1 mm³. There are five different types with different thickness of filters in the capsule of GD-450; namely, 0.2 mm acrylic plate; 0.5 mm acrylic plate; 0.7 mm aluminum filter; 0.2 mm copper filter; and 1.2 mm tin filter. The functions of these filters in

GD-450 are the same as that of SC-1; to estimate radiation energy and to lower the energy dependence effect. The GD-450 dosimeters are the major personal dosimeters used in Japan.
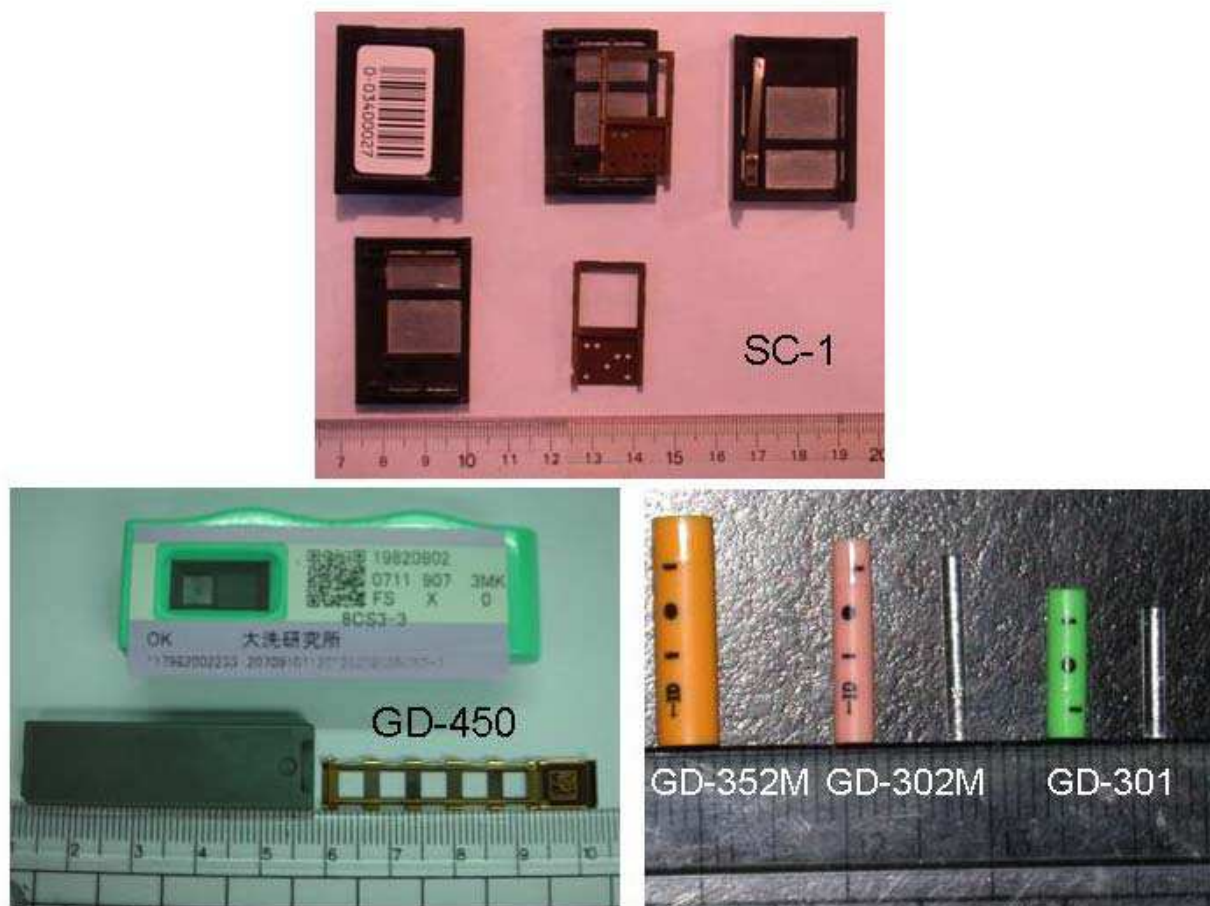

Fig. 7. Three types of RPLGD; above: SC-1 system for environmental radiation monitor; below left: GD-450 system for personal dose monitor; and below right: small volume Dose Ace system for research

The Dose Ace type RPLGD is mainly for research purposes. It is a cylindrical shape with three different models; GD-302M, GD-352M, and GD-301. The GD-302M and GD-352M have a length of 12 mm and a diameter of 1.5 mm, while GD-301 has a length of 8.5 mm and a diameter of 1.5 mm. GD-301 and GD-302M, without filters in capsule, are used to measure the dose of high energy photons as in radiotherapy. However, there is a Tin filter in the capsule for GD-352M to lower the energy dependence effect. The GD-352M can be used for measuring the dose from low energy photons as in diagnostic radiology. In the process of dose readout, based on the dose values, the dose ranges are divided into two categories, low dose range (10 µGy – 10 Gy) and high dose range (1 Gy - 500 Gy).The readout system can automatically distinguish the dose range according to different readout magazine used by the users. On the top of that, there are different readout areas in RPLGD for different dose ranges too. The readout area for high dose range is located at between 0.4 mm and 1 mm, a total length of 6 mm and a total volume of 0.47 mm^3, from the non-series end in the

readout area (as shown in Figure 8); while the low dose range is located from 1 mm to 6 mm with a volume of 0.47 mm^3. The high dose readout area can be used for the measurement of dose with high gradient too. The Table 2 shows the characteristics of various RPLGDs.
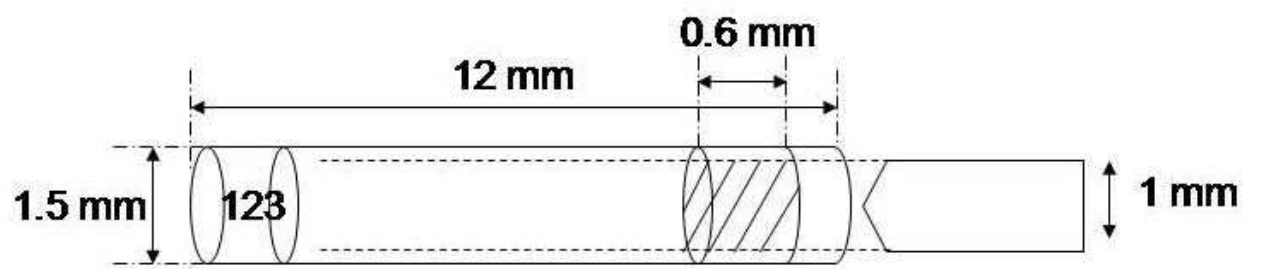

Fig. 8. The high dose readout area for GD-320M; the series end is located on the left side, the readout area is located at 0.4 mm to 1.0 mm from the non-series end, the diameter of incident pulse ultra-violet laser is 1 mm (Hsu).

Type	SC-1	GD-450	Dose Ace
Effective atomic number	12.04	12.04	12.04
The dose linearity range	10 μGy - 10 Gy	10 μGy - 10 Gy	10 μGy - 10 Gy 1 Gy - 500 Gy
Energy dependency（20 keV / ^{137}Cs）	1.2（with energy compensator filter）	1.2（with energy compensator filter）	3.4（w/o energy compensator filter）0.8（with energy compensator filter）
Fading effect	＜ 5 % / yr	＜ 5 % / yr	＜ 5 % / yr
Repeatable readout	yes	yes	yes
Angular dependency	± 8% (0 ~ 80 degree)	± 3% (0 ~ 80 degree)	0 (0 ~ 80 degree)

Table 2. The characteristics of RPLGD

In Figure 9, it shows the readout reproducibility for GD-352M and TLD-100H respectively with a C.V. (coefficient of variation) of 0.46 – 3.11 for GD-325M and C.V. of 0.71 – 3.87 for TLD-100H. The figure shows that the C.V. is smaller for RPLGD as compared to that of TLD because of different manufacture methods. Each RPLGD is made after glass material melted at high temperature and results in a smaller variation among each RPLGD. On the other hand, the TLD is made with growing crystal, therefore the variation is greater.
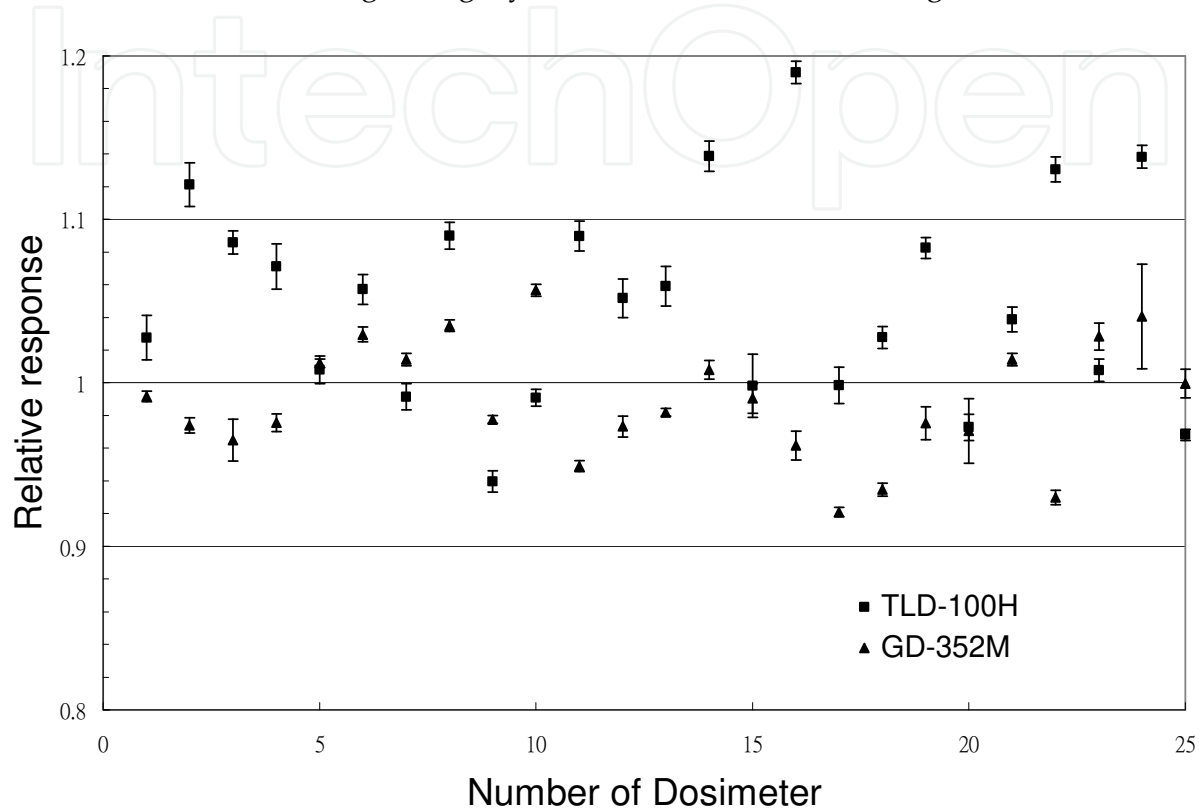

Fig. 9. The readout reproducibility of GD-352M and TLD-100H

Figure 10 shows the dose linearity for GD-352M and TLD-100H respectively in a range of 0.105 mGy and 50.4 mGy. The measured dose points are at 0.105 mGy, 0.168 mGy, 0.672 mGy, 1.05 mGy, 2.1 mGy, 6.3 mGy, 25.2 mGy, and 50.4 mGy with five RPLGDs for each measured point. The correlation coefficient is close to unity for both GD-325M and TLD-100H. It shows that the dose irradiated is proportional to the dose estimated from readout.

Figure 11 shows the energy dependence for GD-302M, GD-352M, and TLD0-100H respectively. The values shown in figure 11 are normalized to the readout from Cs-137 irradiation. When un-filtered GD-302M irradiated with low energy photons, the interactions between photons and RPLGD are increased because of the photoelectric effect. Therefore the luminescence signal is increased too. For filtered GD-352M, the Tin filter can stop the low energy photons; hence, the energy dependence effect is less.

Table 3 shows the characteristics comparisons of different passive dosimeters. It demonstrates that the physical characteristics of OSLD are better than that of TLD. And the physical characteristics of RPLGD are better than that of OSLD because of different readout system and different luminescence material. Therefore, RPLGD could become one of the important dose measurement tools in the future.

	TLD	OSLD	RPLGD
Principe of measurement	luminescence signal	optically stimulated luminescence signal	radiophotoluminescence signal
Luminescence material	crystal	crystal	glass
Excitation source	heat	visible light	ultra-violet laser
Sensitivity	material-dependent	material-dependent	good
Repeatable readout	no	yes, but intensity reduced	yes, with the same intensity
Range of measurement	material-dependent （10μGy - 10 Gy）	material-dependent （10μGy - 10 Gy）	10μGy - 10 Gy 1 Gy - 500 Gy
Geometrical shape	chip and powder	powder	various shapes
Fading effect	material-dependent （5 - 20 % / quarter）	material-dependent （0 - 10 %/year）	less than 5%/year
Energy dependence	material-dependent	material-dependent	± 20% （having energy compensation filter）
Capability to distinguish the types of radiation	yes	yes	yes
Re-useable	yes	no	yes

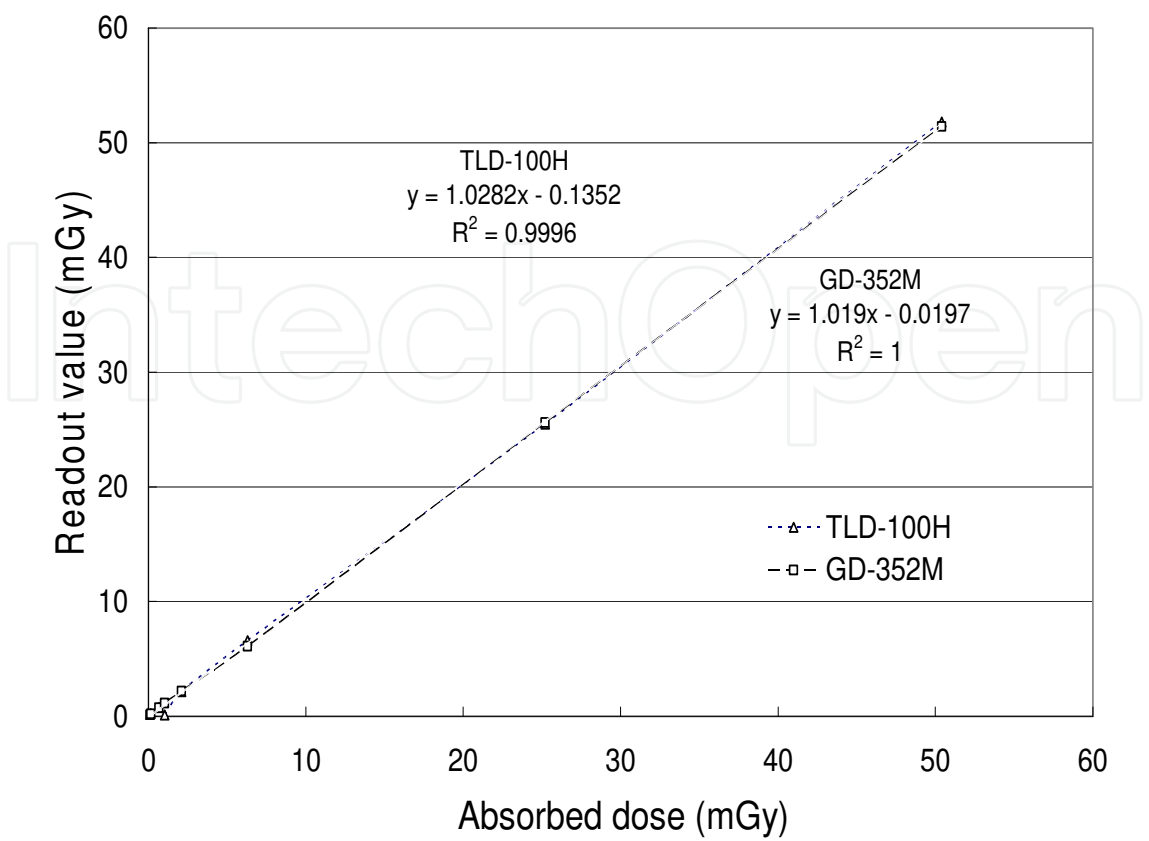Table 3. The characteristics comparisons of TLD, OSLD, and RPLGD

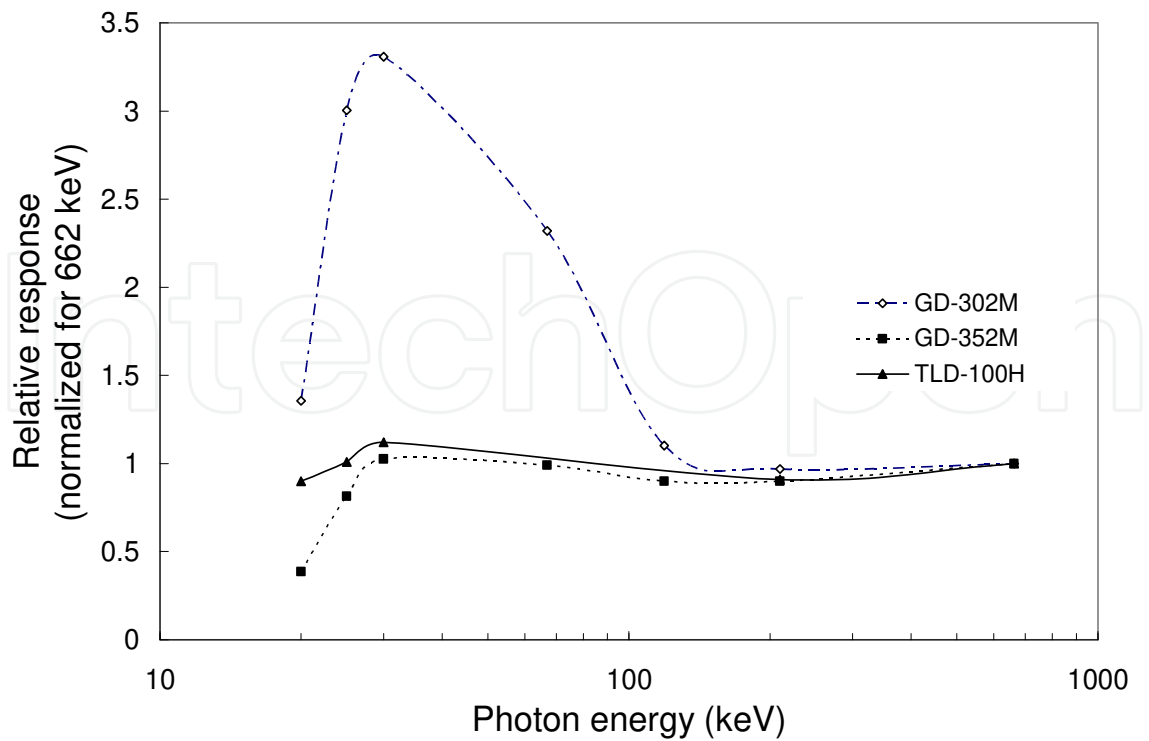Fig. 10. The dose linearity curves for GD-352M and TLD-100H, both C.V.s are less than 3.

Fig. 11. The energy dependence curves for GD-302M, GD-352M, and TLD-100H

7. Characteristics of RPLGD for clinical applications

The clinical applications of RPLGD characteristics are summarized in the followings:
1.   Repeatable readout
The luminescence signal does not disappear after readout; therefore, repeated readout for a single exposure is possible for RPLGD.
2.   Small difference in individual sensitivity
The readout variation between different PRLGDs with the same exposure is small. RPLGD is manufactured with melted glass; therefore, its individual sensitivity is small as compared to that of either TLD or OSLD.
3.   No correction factor needed
The luminescence single can be converted to the exposure dose directly without the need of correction factors. The exposure dose can be determined with the help of readout from reference PRLGD built-in to the readout system.
4.   Small energy dependence
The energy dependence existed in FD-7 glass, if there is no energy compensator filter with it. However, energy dependence can be reduced with energy compensator filter.
5.   Small fading effect
The stability of color centers in RPLGD is high. Hence the effects of environment conditions such as humidity and temperature have very little impact to color centers, hence low fading effects for RPLGD.
6.   Better reproducibility
By using pulse ultra-violet laser as excited source, the accuracy of repeated readout can be maintained. Therefore, RPLGD has a very good reproducibility.
7.   Wide measurable dose range
The dose linearity range for RPLGD is 0 – 500 Gy. This range covers the dose range used in the medical field. RPLGD can therefore be applied for dose verification in radiotherapy as well as in diagnostic radiology. RPLGD is also desirable for high dose gradient area, such as IMRT (Intensity Modulated Radiotherapy) procedures or HDR (High Dose Rate Remote Afterloader) procedures because of its small effective readout area.
8.   Feasibility of personal dose monitor tools
The characteristics, physical and chemical, of RPLGD are equal to or better than that of TLD and OSLD because of its luminescence material and readout technique. Hence, RPLGD can be used as dose monitor for radiation field worker.

8. Applications of RPLGD

Araki applied the RPLGD system in Stereotactic Radiosurgery (SRS) procedure for dose measurements, including Gamma Knife, Cyberknife etc (Araki, Arakia). The results of output factors are comparable with the results from Hi-p Si Stereotactic field detector and Mote Carlo calculation. It shows RPLGD can be used for small field radiation measurements effectively. Nose designed a tube to hold RPLGDs for dose measurements for head and neck patients to verify the delivery dose against the calculated dose from treatment planning system (Nose). Although the maximum dose variation can be as high as 15%; however, those differences are mostly from the positioning errors. Based on the RPLGD physical characteristics study, the error from the RPLGD system stability is less than 3% (out of 15%). Yasuda and Iyogi applied RPLGD in space and environment

radiation monitor (Yasuda, Iyogi). Hsu et al. also applied RPLGD in prostate HDR (High Dose Rate Remote Afterloader) procedure to study the dose distributions (Hsu). Many institutes in US and Europe devote into the developments and the researches in the new luminescence material and readout techniques for RPLGD (Yasuda, Araki, Arakia, Nose, Iyogi, Norimichi ,Hsu).

With its small volume, RPLGD can be used in in-vivo dose measurements; e.g. dose evaluation in animal irradiation study. RPLGD can also be placed in the anthropomorphic phantom to evaluate dose received during the clinical procedures for diagnostic radiology and radiotherapy. With its characteristics of repeatable readout and small effective readout area, RPLGD can also be used in brachytherapy procedures to evaluate the dose delivery accuracy for each procedure as well as for entire course. On the top of that, with the help of dedicated tube to hold RPLGD, one can apply RPLGD in the area of adjacent critical organs to monitor the organ dose to avoid the dose exceeding the tolerance during the radiotherapy procedure. It can improve the patient life quality after radiotherapy.

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Edited by Prof. Hala Gali-Muhtasib

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