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Abstract: The absorbed dose in glass due to electrons or photons can be measured between 10^{-2} and about 10^{8} Gy (10^{0} and 10^{10} rad) using the radiophotoluminescence, the thermoluminescence, and the coloration property of silver-activated phosphate glass. Systematic and statistical measuring errors relevant in practical applications are discussed.

(To be published)

1. Introduction

The measurement of high radiation doses is of practical importance in problems related to radiation damage of materials. In testing material properties under the influence of radiation or in shielding studies to prevent components from getting destroyed by radiation the need arises to measure absorbed doses up to very high values. For example, in high-energy electron accelerators the beam energy and/or beam power can be so high that radiation damage of machine components (electrical insulators, water hoses etc.) occur due to the field of scattered electrons and photons. A special problem is the production of synchrotron radiation with critical energies higher than 25 keV in high-energy circular accelerators; with these energies the synchrotron radiation reaches the energy range of soft x-rays and penetrates the vacuum chamber partly, giving rise to high radiation doses in the accelerator room. In such cases a permanent survey of the doses at many points along the large machine components is desirable.

Solid state dosemeters are suitable for these purposes. They should have the following properties: It should be possible to interprete the measured magnitude in terms of a dose absorbed in a mean accelerator material; the energy range in which the calibration of the dosemeter is valid, should be known and should extend to possibly low photon energies; in cases where the field is strongly absorbed by machine parts the doses may change drastically from one point to another, therefore the dynamical range should comprise many orders of magnitude; for a long-term survey of irradiated materials a small fading of the dosemeter is necessary; sometimes the number of measuring points is large, therefore the dosemeters should be cheap, rugged and easy to handle.

These requirements are fulfilled in a nearly ideal way by silver-activated phosphate glass dosemeters.

2. General properties of silver-activated phosphate glass

In the Sixties numerous articles were published about glass dosimetry, but in the last decade only few papers with new results appeared. Good reviews can be found in refs. 1 and 2. In the following some properties of glass dosemeters will be mentioned which are useful for routine measurements of high doses.

The composition of a typical silver-activated dosemeter glass is given in table 1. From these data and the known elemental photon cross sections the mass energy-absorption coefficients can be calculated; it is shown in fig. 1 as a function of photon energy, together with the corresponding coefficient of aluminium.

Both curves are very similar, and also the glass density (2.6 g cm^{-3}) is similar to that of aluminium. Therefore the physical magnitude measured with such a dosemeter glass can be regarded as the absorbed dose in aluminium which is a typical material of an accelerator component (e.g., the building material of the vacuum chamber). The size of the glass dosemeter which we use for all our measurements is 1 mm ∅ x 6 mm length. This size determines the photon energy range in which true measurements of absorbed dose in aluminium are possible or, in other words, where the ratio of absorbed dose to kerma is unity. Deviations from unity are expected at low energies, where photons are absorbed in the surface layer (and in a plastic cover), and also at high energies where Compton electrons will escape. Yamaguchi (ref. 3) has calculated the useful range, the results are also indicated in fig. 1 for a glass covered with 1.5 mm of plastic or 1.5 mm of aluminium. The energy range from 20 keV to 2 MeV or from 30 keV to 3.5 MeV is sufficiently large for all practical purposes.

The glass property which has been used up to now exclusively for reading the deposited energy in practice is the radiophotoluminescence (RPL). Stable fluorescence centers are created by ionizing radiation which emit light of about 600 nm wave length when, in a reading instrument, excited by UV light of 360 nm. The minimum detectable

radiation dose is limited by the background fluorescence of the glass, the "predose", and by some fraction of the low-energy tail of the UV spectrum scattered into the photomultiplier. For a fresh batch of our 1 mm ∅ x 6 mm rods the mean predose was found to be equivalent to 0.015 Gy*) with a standard deviation of 40%. After repeated use and heat treatments for thermal regeneration the predose increases up to approximately 0.03 Gy.

The RPL information stored in the glass is very stable. After the end of a short irradiation a 10% increase of the signal is observed. This process is finished after approximately 20 h, so for accurate measurements a waiting time of one day is necessary. A long-term decrease (fading) of the RPL greater than 10% has not definitely been established, if the dosemeter is stored at room temperature (see ref. 1). We can assume that the signal is stable for a time period of several years. The stability of color centers produced by high doses is discussed in section 3.

After an irradiation and measurement a thermal treatment at 400° C regenerates the dosemeter. The heating time we use to erase all information depends on the accepted dose: 1.5 h for doses lower than 10 Gy; 4 h for 10 to 10^3 Gy; 7 h for 10^3 to 10^7 Gy. If the absorbed dose is higher than 10^2 Gy the color centers formed by the ionizing radiation become visible, the color of the glass changes from light yellow at 10^3 Gy to black at 10^8 Gy. After the mentioned annealing the glasses having received doses as high as 10^7 Gy are completely clear again, only their predose increases in a rather irregular way. A rough indication of measured predoses are given in table 2. This increase of the predose restricts the reusability to a certain extent, another restriction is mentioned in the next section.

*) 1 Gray (Gy) = 100 rad

The glass rods are cheap (2 DM per piece*)) and easy to handle, therefore large quantities can be used for special purposes. They can be cleaned simply by stirring in methanol. Only if the dose to be measured is comparable with the predose and subtraction of both quantities is necessary, a successive washing in acetone, running water, destilled water and methanol is advisable before measuring both predose and dose.

3. Measuring range and accuracy

There is a strictly linear relationship between absorbed dose and the RPL signal for doses up to 50 Gy. In this dose range we obtain with our reading instrument**) the following accuracies. For doses well above the predose and with a new batch of glass dosemeters the standard deviation is 3%. After frequent use and many heating cycles, especially after frequent measurement of high doses, the standard deviation increases up to 6%. This may restrict the reusability of the dosemeters in cases of very accurate measurements. To these mainly statistical errors the error due to the long-term stability of the reader has to be added which is 1 to 2% in our case. Therefore the total measuring accuracy of a dose measurement between 0.5 and 50 Gy with, say, 10 dosemeters to reduce the statistical error is less than the usual accuracy of the dose rate of the radioactive source used for calibration (mostly 5%), and it is much smaller than the systematic errors inherent in the measurement of absorbed doses due to the difference between glass and the material in question, lacking radiation equilibrium etc. It should be mentioned that silver-activated phosphate glasses from different manufacturers or different batches from the same company may have different RPL sensitivities (and TL sensitivities, see below); we observed differences up to a factor of 1.5.

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*) Schott Jenaer Glaswerke, Mainz, Germany

**) FGD6, Tokyo Shibaura Electric Co.

For doses below 0.5 Gy the predose of a dosemeter must be subtracted and the standard deviation of a series of measurements increases, our results are indicated in table 3. So the lowest dose detectable with the glass rods is roughly 0.02 Gy.

Absorbed doses higher than 50 Gy will change the color of the glass. The color centers first will attenuate the stimulating UV light so that, above 10^3 Gy, the fluorescence decrease with increasing dose. For doses higher than 10^5 Gy also the emitted red light is attenuated and the output signal drops even faster with dose, until the glass becomes more or less black. Freytag (ref. 4) already pointed out that also this decreasing branch could be used for dose measurements, provided that a well-determined calibration curve is available. We received the necessary calibration curve for doses up to $5 \cdot 10^4$ Gy by means of a ^{137}Cs source (activity 6 Ci), for higher doses we used the electron-photon stray radiation very near to the converter target of our linear accelerator where the positrons are produced by 250 MeV electrons. The dose rate of this field varies between 10^2 and 10^5 Gy/h as a function of distance. We first checked that the special field distribution is constant during routine use of the accelerator. Then we determined the calibration curve in an iterative way: the irradiation time was chosen such that the glasses in most of the measuring positions received doses which lay inside the range of the already determined part of the curve and that some glasses received higher doses which lay outside. The result is shown in fig. 2. Also indicated are the glass colors to distinguish between the two branches of the curve. The accuracy of the calibration is 20% in the range 10^2 to $5 \cdot 10^4$ Gy and 25% between $5 \cdot 10^4$ and $2 \cdot 10^7$ Gy. The glass rods show no systematic change in sensitivity larger than 5% after receipt of doses as high as $7 \cdot 10^7$ Gy and after the appropriate heat treatment, only the predose becomes larger (see section 2) and the standard deviation of the next dose measurement has somewhat increased due to induced statistical irregularities.

For the use of the decreasing branch of the calibration curve it is important that the produced color centers are stable for long time periods, since measured high doses are often the result of long measuring times. A fading of the optical density during the first few days after a short irradiation was observed by Becker (see ref. 1). We checked the stability of dose readings over a period of 60 days after an irradiation time of 4 days and an additional waiting time of 1 day; the dose range was $7 \cdot 10^3$ to $4 \cdot 10^6$ Gy. No change in the dose reading larger than 8% was observed for doses below $1 \cdot 10^6$ Gy. Short-term variations could not be measured for such high doses, but they are of minor practical importance and are roughly accounted for by using the calibration curve (fig. 2) which also was determined by long-term irradiations. In the range $1 \cdot 10^6$ to $4 \cdot 10^6$ Gy, where the glasses show already a dark brown color, we observed a decrease of the dose reading of 20% within the first 30 days; for the next 30 days the glasses roughly remained stable.

From fig. 2 it is clear that no dose measurement is possible by using the RPL in the range $2 \cdot 10^2$ to $2 \cdot 10^3$ Gy. This gap can be filled by using the thermoluminescence (TL) property of silver-activated phosphate glasses. Radiation-induced TL has already been mentioned by Regulla in ref. 2. Using a linear heating rate he found four TL peaks at the following temperatures: 25^o (blue light), 115^o (red), 165^o (blue) and 230^o C (red). We used a commercial TL reader*) and in the beginning an (unlinear) heating rise up to 320^o within 15 s. Not all of the TL centers were emptied in this way, another heating cycle gave an output 5% of the first cycle, the details depend on the reading instrument. Then we noticed that an appreciable part of the light is emitted by the low-lying centers which give severe fading of the total output (30% after 10 days). Therefore we finally used two successive cycles: preheating at 150^o C in 15 s and light integration at 320^o in 15 s. With these parameters no fading greater than 5% was observed during a time period of 90 days.

*) TLR6, Eberline Instrument Corporation

After the end of an irradiation a waiting time of 1 day at room temperature is necessary, the same precaution as in the case of a RPL measurement. It can be shown that part of the absorbed energy is first trapped in low-lying centers and then, in the course of several hours, is transfered to centers which will be emptied at higher temperatures; therefore a varying amount of light will get lost during the preheat cycle if applied immediately after the irradiation.

A calibration curve for the TL signal was taken in the same way and with the same accuracy as mentioned above, it is also shown in fig. 2. Only the linear part is interesting, it covers the range where a dose measurement by RPL is not possible. The glasses show no background reading (TL predose), therefore the lowest detectable TL signal depends on the background of the reading instrument; in our case the zero reading was equivalent to a dose of about 15 to 30 Gy. The standard deviation of a series of measurements was 10% at 10^2 Gy and 8% at 10^3 Gy. The sensitivity of a glass dosemeter as defined by the linear part of the curve may vary by a factor of 2 from one batch of dosemeters to another received from the same manufacturer, and this variation is not proportional to the variation in RPL sensitivity. Therefore at least one calibration point is necessary for each batch.

All TL information is completely annealed by the heat treatment mentioned above. A dependence of the TL sensitivity on the heating cycle, especially on the cooling rate as observed with other TL dosemeters like LiF, could not definitely be established. No change of sensitivity and no increase in the standard deviation is observed after receipt of doses as high as 10^7 Gy and after the appropriate annealing. After received doses between 10^7 and 10^8 Gy we observed an irreversible increase of the sensitivity by a factor of 1.5 which restricts the reuse of the glass dosemeter.

4. Literature

1. J. F. Fowler and F. H. Attix, Solid state integrating dosemeters. E. Piesch, Developments in RPL dosimetry. Both articles in: F. H. Attix (ed.), Radiation Dosimetry, Vol. II (1966) and Suppl. 1 (1972), Academic Press, New York

2. D. F. Regulla, Internal Report S 466 (1977), Gesellschaft für Strahlen- und Umweltforschung, München

3. C. Yamaguchi, Nucl. Instr. Meth. 197 (1982) 473

4. E. Freytag, Health Physics 20 (1971) 93

Tables

Table 1 Elemental composition of silver-activated phosphate glass

Element	Percentage by weight
O	53.6
P	33.2
Al	4.7
Ag	4.1
Li	3.5
B	0.9

Table 2 Measured predoses after receipt of high doses and annealing

Dose before annealing (Gy)	Predose after annealing (Gy)
$< 10^3$	0.03
$1 \cdot 10^4 - 5 \cdot 10^5$	0.1 - 0.3
$5 \cdot 10^5 - 1 \cdot 10^7$	0.3 - 1.5
$1 \cdot 10^7 - 1 \cdot 10^8$	$\sim$ 2

Table 3 Standard deviation σ of low dose measurements with glass dosemeters of 1 mm Ø x 6 mm length

Dose (Gy)	σ (%) with new dosemeter	σ (%) with dosemeter after repeated use
1	3	5 - 7
0.1	5	10
0.04	9	19
0.02	12	26
0.01	(40)	(60)

<u>Figure Captions</u>

Fig. 1 Mass energy-absorption coefficients of silver-activated phosphate glass (full line) and aluminium (dashed line), see left scale. Right scale and dashed-dotted lines: ratio of absorbed dose to kerma for a glass dosemeter 1 mm ∅ x 6 mm length covered with 1.5 mm of plastic (a) or with 1.5 mm of aluminium (b).

Fig. 2 RPL and TL calibration curve for glass dosemeters 1 mm ∅ x 6 mm length (Manufacturer: Toshiba corporation, Tokyo, Japan)

Fig. 1

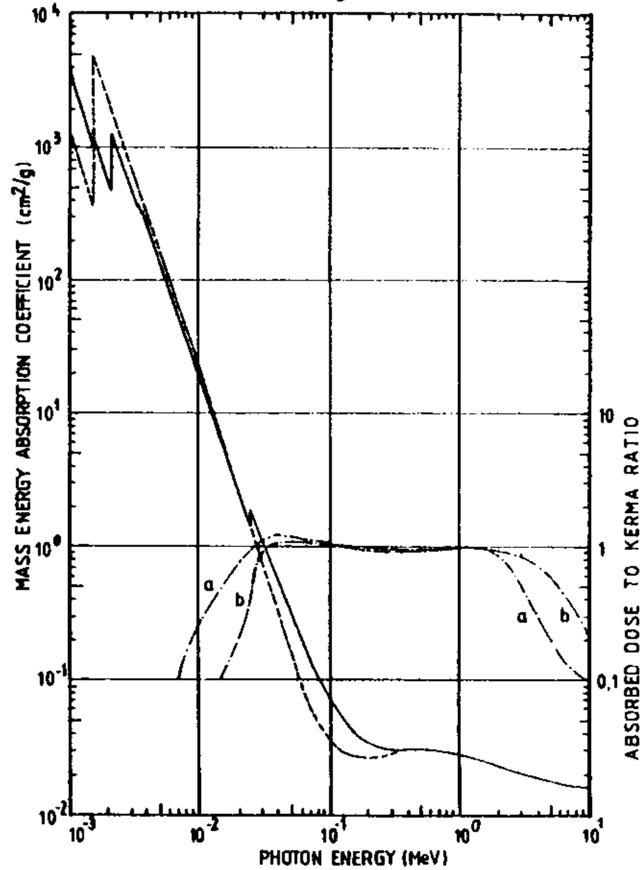
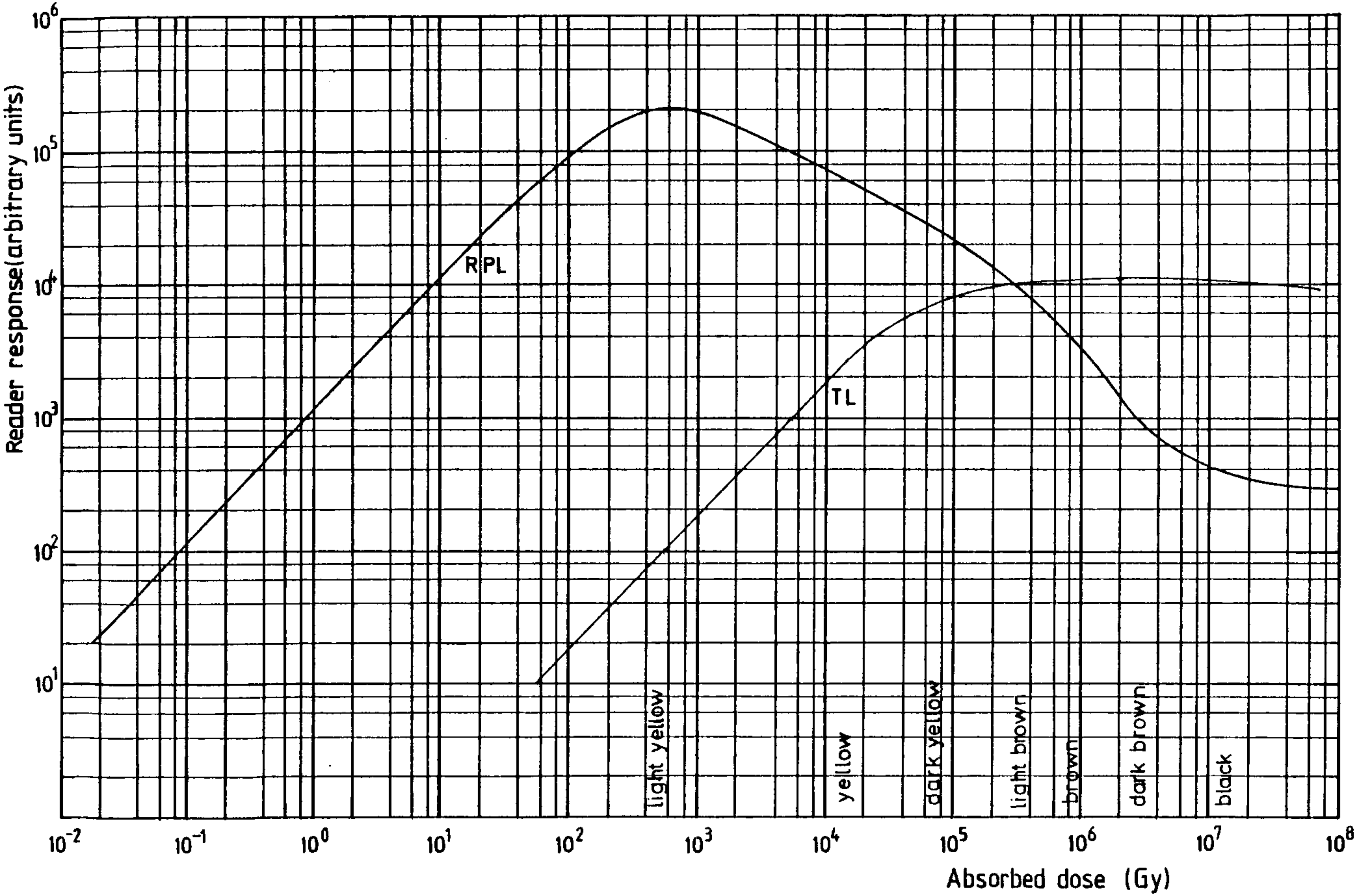


Fig. 2



Reader response (arbitrary units)
10^6
10^5
10^4
10^3
10^2
10^1
RPL
TL
light yellow
yellow
dark yellow
light brown
brown
dark brown
black
10^{-2}
10^{-1}
10^0
10^1
10^2
10^3
10^4
10^5
10^6
10^7
10^8
Absorbed dose (Gy)