Title page

a) Title of article

Evaluation of the basic properties of the BANGkit™ gel dosimeter

b) Authors and addresses

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c) Short title

Basic properties of the BANGkit™ gel dosimeter

d) Keywords

polymer gel, gel dosimetry, BANGkit

Abstract

We evaluated the basic properties of a commercially available BANGkit™ gel dosimeter, which is a normoxic type of BANG® gel. This gel-kit has the same composition as the BANG®3 gel, but is fully oxygenated. To exclude oxygen, oxygen scavenging ascorbic acid and copper sulfate as a catalyst are used. The properties that we examined are the effects of the concentrations of copper sulfate and ascorbic acid on the response, the reproducibility, the long-term stability, the temperature effect at irradiation, and the dose-rate effect. In our results, the excellent linear fit of the R2-dose response in a dose range for clinical use and its reproducibility were observed. The precision of a linear fit was preserved for about three weeks. The temperature at irradiation showed a significant effect on the dose response. Although the dose-rate dependence in the high dose range was observed, it was negligible for the clinical dose range up to 270 cGy. In conclusion, this gel dosimeter is thought to be utilizable in clinical practice, while we have to pay attention to the temperature during the entire measurement processes, and additionally there is room for improvement in the linearity and the dose-rate dependence in the high dose range.

1. Introduction

Due to the significant progress of radiation therapy equipment and radiation treatment planning system, it is now possible to concentrate a high dose to the target and to decrease the dose to the organ at risk. Clinically, three dimensional conformal radiation therapy, intensity modulated radiation therapy and stereotactic radiosurgery (radiotherapy) have been implemented in clinic. For quality assurance of these complicated radiation therapy technologies, a three-dimensional dosimetry system with high spatial resolution and high sensitivity is necessary. Conventional dosimeters such as ionization chambers, thermoluminescent dosimeters and silicon diodes measure a point dose (one-dimension). Radiographic films offer two-dimensional dose measurement.

Polymer gel was developed as a three-dimensional dosimeter. The foremost property of polymer gel dosimeters is that ionizing radiation produces free radicals in both the solvent (such as water) and the solutes (such as the gelling agent or the acrylic monomers), in proportion to the absorbed dose, and these in turn initiate free radical chain polymerization reaction that leads to the formation of polymer microparticles which remain attached to or entangled with the gelling substance. Therefore, the spatial distribution of the polymer in the gel represents the dose distribution of the radiation field in the gel (Maryanski *et al* 1993, 1994a, 1999). Since the development of the first polymer gel dosimeter by Maryanski *et al* (1993), various gel compositions have been proposed and a lot of investigations have been done to evaluate the dosimeters.

The response of polymer gels is sensitive to oxygen, as oxygen is an inhibitor of the radiation-induced polymerization (Maryanski *et al* 1993). One of the problems in the

handling of the polymer gel dosimeter was the difficulty in manufacture, particularly the need to exclude oxygen both from the gel and the manufacturing process. This is achieved by bubbling inert gas heavier than oxygen (e.g. nitrogen) through the gel solutions. Use of the oxygen scavengers was considered to solve this problem (Maryanski *et al* 1994a), and several reports of experiments with this so-called normoxic gel have been published recently (Bayreder *et al* 2006, De Deene *et al* 2002a, 2006, Fong *et al* 2001, Hurley *et al* 2005, Maryanski *et al* 2002). This normoxic polymer gel can be made under normal atmospheric conditions and its manufacture is relatively simple and convenient.

BANG® gel (MGS research Inc., Guilford, CT, USA) is one of the most extensively studied polymer gels. So far three types of gel (BANG®1, BANG®2, and BANG®3) have been proposed sequentially and the properties of the gels have been improved in dose range, sensitivity and toxicity (Maryanski *et al* 1993, 1994b, 1996, 1997, 1999). In this work, we examined a commercially available BANGkit™ gel dosimetry system, which is a normoxic type of BANG gel. The BANGkit™ gel has the same composition as the regular BANG®3 gel (gelatin + methacrylic acid (6% by weight) + water), but it is fully oxygenated. Therefore, it does not respond to radiation unless the dissolved oxygen is thoroughly removed from it. In order to de-oxygenate the gel, oxygen scavenging ascorbic acid and copper sulfate ($CuSO_4$) as a catalyst are used. Advantages of the BANGkit™ gel dosimeter are that we are able to manufacture the gel whenever it is needed as well as to easily change the container with different shapes.

The purpose of this study was to evaluate the basic properties of BANGkit™ gel dosimeter. We studied the effects of the concentration of ascorbic acid and copper sulfate on the response, the reproducibility, the long-term stability, the temperature

effect at the time of irradiation and the dose-rate effect.

2. Materials and Methods

2.1 Gel preparation

Procedures of gel preparation were as follows. BANGkit™ was contained in a 500 ml flat-wall polystyrene flask and all components were stored in a refrigerator until the day of gel preparation. Beforehand, we made a 1-millimolar-batch solution of copper sulfate by dissolving the copper sulfate powder in deionized distilled water. All vials for experiments were filled with an oxygen-scavenging solution of the same concentration as the one used for making the gel two days before gel preparation. On the day of gel preparation, the gel in the flasks was melted in a hot water bath at about 55 ^{o}C. After the flasks had been in the hot water bath for 1 hour, the melted gel was poured into the mixing container. The mixing container was placed back in the hot water bath for 1 hour. Then the ascorbic acid solution was added and the sol was rapidly mixed with the overhead mixer for 5 minutes. Next the copper sulfate solution was added and the sol was mixed for 10 minutes. The oxygen-scavenging solution was removed from the vials and the sol was poured into the vials. The sol in the vials was left for gelation for two days. Irradiation of the gel was performed on the third day and irradiated vials were left for one day in the MRI room after irradiation in order to bring the gel temperature to that of the MRI room temperature. The gel was imaged on the fourth day (except the experiment for stability). We used 2.0cm diameter and 15cm long cylindrical Pyrex

vials for this work. The entire process of the preparation of the gel was performed in normal room atmosphere and under the safe-light lamp.

2.2 Irradiation

The gel-filled vials were placed one at a time in a water tank (30cm x 34cm x 37cm) with their longitudinal axis perpendicular to the beam central axis. The vials were irradiated with an opposing portal irradiation by 10 x 10 cm^2 fields using 6 MV photons of a Varian Clinac 2300 C/D linear accelerator. The individual doses of the vials were calculated from the dose data measured by an ionization chamber (N30001, PTW Freiburg, Germany) placed at the same location as the vials for experiments beforehand.

2.3 MRI scan and data analysis

MRI scans were carried out using a 1.5T GE SIGNA Horizon MRI scanner. A spin-echo (SE) pulse sequence was used with the repetition time (TR) of 2 s and echo times (TE) of 20 and 100 ms. Other scanning parameters were as follows; slice thickness = 5 mm, field of view (FOV) = 24 cm, matrix size = 256 x 256, the number of acquisitions (NEX) = 2 and using 'Interleave' option.

It is known that an MR imaging pulse sequence with a larger number of TEs (e.g. CPMG) leads to higher precision of measured T2 values (De Deene and Baldock 2002b). There are two reasons not using CPMG. First is its availability, in particular, of an MRI protocol using 16-32 echoes without significant signal artifacts. Second, when spin-echo signals are taken by averaging over a large volume such as for the current

study, one can obtain sufficiently accurate T2 values using the two-point SE sequence. The accuracy is sufficient for studies to evaluate the parametric dependence of measured R2 values.

The MR images were transferred to a personal computer for further analysis. The transverse relaxation rate R2 (1/T2) of the water protons was taken as a measure of radiation dose absorbed in the gel. For calculation of R2, MATLAB software (The Math Works Inc., Natick, MA) was used.

2.4 Experiments

2.4.1 The effects of the concentrations of copper sulfate and ascorbic acid

The concentrations of ascorbic acid and copper sulfate in the gel were coded with two numbers following the words “AC”. The first number indicates the millimolar concentration of ascorbic acid and the second number indicates the micromolar concentration of copper sulfate. For example, AC1-5 means that 1 millimolar of ascorbic acid and 5 micromolar of copper sulfate are dissolved in a litre of the gel. In this experiment, we manufactured 12 different concentrations of gels as follows; AC0-0, AC0-5, AC1-5, AC1-15, AC1-30, AC2-10, AC2-30, AC2-50, AC3-15, AC3-30, AC4-20 and AC6-30. These gel vials were irradiated with doses ranging from either 0-20 Gy or 0-80 Gy.

2.4.2 Reproducibility

To evaluate the reproducibility, we measured the dose response of the AC1-5 gel four times separately. Gel 1 and Gel 2 were made from the same gel kits, but at a different time. Gel 1 was made one month after receiving gel kits and Gel 2 was made 4 month after receiving gel kits. Gel 3 and Gel 4 were made from another gel kits, and Gel 3 and Gel 4 were made 2 and 6 month after receiving gel kits, respectively. The gels were irradiated with a dose range of 0-12 Gy (Gel 1-3) or 0-10 Gy (Gel 4).

2.4.3 Stability

Evaluation of the stability after irradiation was performed using AC1-5 gels irradiated with a dose range of 0-80 Gy. MRI scan was performed on Day 1, 2, 3, 4, 6, 11, 16 and 23 post-irradiation. The gels were placed in the dark at MRI room temperature during this experiment.

2.4.4 Temperature effect at the time of irradiation

We used the AC1-5 gels for this experiment. The temperature of the gels at the time of irradiation was adjusted to 5.6, 15 and 25 ^{o}C using an automated cooler-heater circulator (Zensui ZR130, Osaka, Japan). Temperature change during irradiation was within ±0.3 ^{o}C. The gel vials were irradiated with a dose range of 0-20 Gy at each temperature. We prepared one gel-filled vial, in which an alcohol thermometer was set, and confirmed that the gel temperature reached the same temperature as the water in the water tank.

2.4.5 Dose-rate effect

For evaluation of the dose-rate effect, we prepared 5 sets of AC1-5 gels and they were irradiated with a dose range of 0-10 Gy with a dose-rate of 68, 136, 204, 272 and 408 cGy/min. Separately, we prepared 2 sets of AC1-5 gels and the gel vials were irradiated in 0.3 Gy steps between 0-2.7 Gy with a dose-rate of 136 cGy/min and 408 cGy/min to investigate the dose rate effect in the lower dose range in more details.

3. Results

3.1 The effects of the concentrations of copper sulfate and ascorbic acid

The R2-dose response curves of the gels up to 50 Gy with various concentrations of ascorbic acid and copper sulfate are shown in Figure 1a. The AC0-0 gel and AC0-5 gel without oxygen scavenger, ascorbic acid, showed no response to irradiation. On the other hand, all gels in which ascorbic acid was added showed response to irradiation. Figure 1b focuses on the linear dose response region of the AC1-5, AC2-10, AC3-15, AC4-20 and AC 6-30 gels. In this experiment, we found that when we increased the concentration of ascorbic acid and copper sulfate of the gel with the same mixture ratio of AC1-5 gel (e.g. AC2-10 or AC3-15 gel), the slope of the R2-dose response curve decreased and the dose range of a good linear fit widened to higher dose (Figure 1b). AC1-5 gel, AC2-10 gel and AC3-15 gel showed a good linear fit in the dose range 0-16 Gy (r^2=0.9999), 0-30 Gy (r^2=0.9985), and 0-40 Gy (r^2=0.9980), respectively. With much more increase of their concentration, we observed an increase of the background R2 value and the R2-dose response curves of AC4-20 gel and AC6-30 gel in the higher

dose range did not maintain a precise linear fit compared with AC1-5, AC2-10 and AC3-10 gels (Figure 1b). When the mixture ratio of ascorbic acid and copper sulfate was different from that of AC1-5 gel, sometimes the deviation from linearity was observed (Figure 1a).

3.2 Reproducibility

Figure 2 shows the R2-dose response plots, measured in four separately prepared AC1-5 gels. The separate linear-fits for Gels 1, 2, 3 and 4 were described by the equations; R2 =3.122 + 1.226D, r^2 = 0.9981; R2 =2.661 + 1.144D, r^2 = 0.9996; R2 =2.933 + 1.180D, r^2 = 0.9996; R2 =2.979 + 1.284D, r^2 = 0.9986 (D= dose in Grays). The means of R2(0) and the slope are 2.924±0.193 and 1.209±0.061, respectively. Considering the statistical uncertainty of the R2 measurements with the current method, we think that the reproducibility of the linear fits of AC1-5 gels in the dose range of 0-12 Gy is good.

3.3 Stability

Figure 3a shows the R2-dose response plots at Day 1, 2, 3, 4, 6, 11, 16 and 23 post-irradiation of AC1-5 gels. In the dose range of 0-16 Gy, the linear fit of each day was stable during day 1 to day 23 post-irradiation (r^2=0.9994-0.9999, Figure 3b). The R2-dose response of the gel was found to be relatively stable after day 4 post-irradiation, although slight changes in the R2 (0) value and the slope of the linear dose response curve were found with increasing time (Figures 3c and 3d).

3.4 Temperature effect at the time of irradiation

Figure 4a shows the R2-dose response plots for AC1-5 gels obtained when the gel temperature at the time of irradiation was 5.6, 15 and 25 ^{o}C. The temperature at the time of irradiation showed a significant effect (the slope of the R2-dose response curve increased from 1.378 $s^{-1}Gy^{-1}$ at 25 ^{o}C to 1.763 $s^{-1}Gy^{-1}$ at 5.6 ^{o}C (Figure 4b)).

3.5 Dose-rate effect

Figure 5a shows the R2-dose response plots for AC1-5 gel when the dose rate at D_{max} varied from 68 to 408 cGy/min. The dose-rate dependence of R2-dose response was observed in the medium (3-8 Gy) and high dose region (8-10 Gy). Figure 5b shows the dose response plots of the gels irradiated in 0.3 Gy steps up to 2.7 Gy with a dose-rate of 136 cGy/min and 408 cGy/min. The R2-dose response plots of these gels were almost identical and this result suggests that there is no significant dose-rate dependence in the low dose region. Figure 5b also shows a good linear fit of R2-dose response in the low dose range. The linear-fits of the gels irradiated with a dose-rate of 136 cGy/min and 408 cGy/min were described by the equations; $R2 = 1.797D + 3.374$, $r^2 = 0.9984$ and $R2 = 1.752D + 3.428$, $r^2 = 0.9993$, respectively.

4. Discussion

Fong *et al* proposed the normoxic gel as the acronym MAGIC (Methacrylic and Ascorbic acid in Gelatin Initiated by Copper) gel in 2001. In their gel system, copper

sulfate and ascorbic acid were added to form a complex with oxygen, and to serve as a free radical source for the initiation of the polymerization of methacrylic acid. Further investigations have been performed into the fundamental properties of the normoxic gel by several authors (De Deene *et al* 2002a, 2006, Gustavsson *et al* 2003, Hurley *et al* 2005, Maryanski *et al* 2002). In these reports, the linear fit of the dose response curve of normoxic gel was not discussed in detail. The gradient of the quasi-linear increase of the R2-dose response curve is generally used in polymer gel dosimetry, although a divergence from linearity has been repeatedly observed (De Deene *et al* 2000, Ibbot *et al* 1997, Oldham *et al* 1998). Meanwhile the advantages of linear equations over nonlinear equations was reported recently (Watanabe 2005), and the author proposed the linearization methods. In our study, we investigated the dependence of the concentration of ascorbic acid and copper sulfate on the dose range with a linear fit of the R2-dose response curves in BANGkit™ gel dosimetry system. Without ascorbic acid, no response to the irradiation was observed with or without copper sulfate. AC1-5 gel showed an excellent linear fit in the dose range of 0-16 Gy. In addition, a good linear fit in the lower dose area (up to 2.7 Gy) was confirmed. With increase of the concentration of copper sulfate and ascorbic acid with the same mixture ratio as AC1-5 gel, a good linear fit up to higher doses was observed (0-30 Gy in AC2-10 gel and 0-40 Gy in AC 3-15 gel). With much more increase of their concentration (AC4-20 and 6-30), the R2-dose response curves in the higher dose range did not maintain a precise linear fit compared with AC1-5 gel, AC2-10 gel and AC3-10 gel. When the mixture ratio of ascorbic acid and copper sulfate was different from that of AC1-5 gel, sometimes the deviation from linearity was observed. The reasons for this are possibly due to the catalytic effect by the mixture ratio of ascorbic acid and copper sulfate or due to the

effect of time needed to achieve de-oxygenation. Further investigations on these factors are necessary.

In the experiments for the reproducibility, we examined the dose response from different gel kits, each of which was prepared at a different time. We found a good linear fit up to 12 Gy in every gel (Figure 2). This result suggests that the BANGkit™ gels before gel preparation with oxygen scavenger can be preserved in the refrigerator for several months.

The long-term stability of the dose distribution of the polymer gel is possibly affected by several factors such as the intercept of the dose-R2 plot due to the gelation process (De Deene *et al* 2000), the spontaneous polymerization by the impurities in gelatin and the degradation of the gel (Maryanski 1999). From our results, the R2-dose sensitivity tended to decrease with increasing time and became almost stable after day 4 post-irradiation. It may be difficult to mention this result only from the point of view of the stability with disregard to some sort of effects of each MRI scan. However the precision of a linear fit of the R2-dose response curve was preserved during the period of this experiment. We think that the dose distribution in the gel can be evaluated for several weeks after irradiation, however both of the gel phantom and the calibration vials should be scanned together.

Few reports have been published about the effect of temperature at the time of irradiation. Maryanski *et al* reported that the BANG®1 gel's (Bis, Acrylamide, Gelatin, Water, Nitrogen-bubbled) dose response did not depend on the temperature at irradiation in 1997. In our study we found the temperature dependence of R2-dose sensitivity during irradiation in the BANGkit™ gel dosimeter. This is most probably related to different kinetics of polymerization chain propagation and termination due to

the difference of chemical compositions between the BANG®1 gel system and the BANGkit™ gel system. Salomons *et al* (2002) reported that the temperature of the gel changed in proportion to the absorbed dose in the gel during irradiation. In their article, they described that it was caused by radiation-induced exothermic polymerization reactions. De Deene *et al* (2006) reported that in comparison to polyacrylamide-based normoxic dosimeter (nPAG), methacrylic acid-based normoxic dosimeter (nMAG) showed a much larger temperature dependence. The BANGkit™ gel is methacrylic acid-based normoxic gel; hence, our results are consistent with these results. The temperature effect during MRI scanning on the R2 values of polymer gel dosimeters was described in many previous studies (De Deene *et al* 1998, 2006, Maryanski *et al* 1994b, 1997, Spevacek *et al* 2001). Although we do not show our result in this article, we evaluated this effect and confirmed the same results as their reports. It is important that the temperature of gel dosimeters is kept constant prior to MRI and that the experimental gel phantoms are scanned together with the calibration vials.

The reports of no dependence on dose-rate in the BANG®2 polymer gel dosimeter have been published by a number of groups (Maryanski *et al* 1996, Novotny *et al* 2001). However, in the normoxic polymer gel dosimeter, several groups have recently reported the dose-rate dependence on the dose response (Bayreder *et al* 2006, De Deene *et al* 2006). Bayreder *et al* reported about the properties of the normoxic polymer gel with tetrakis-hydroxy-methil-phosphonium chloride (THPC) as an oxygen scavenger, and they found the dose response dependence on dose rate in the medium and high dose regions and no significant dependence in the low dose level. Our results were consistent with these results. From our results the dose-rate independence for the clinical external beam dose range up to 270 cGy is negligible.

5. Conclusion

In this work, we have demonstrated the basic properties of the BANGkit™ gel dosimetry system. The preparation of gel is easy and does not take long time. The excellent linear fit of the R2-dose response in the dose range for clinical use and its reproducibility were observed. The precision of a linear fit of the R2-dose response curve was preserved for about three weeks. The temperature at the time of irradiation showed a significant effect on the dose response and we must pay attention to the temperature during the entire measurement processes. Additionally, although the dose-rate dependence was observed in the high dose range, i.e. 8 Gy and higher, it was negligible for the clinical dose range up to 270 cGy. We conclude that this BANGkit™ system is utilizable in clinical practice, although there is room for improvement in several aspects, which include the linearity of response in the higher dose range, in particular, for stereotactic radiosurgery applications and the dose-rate independence in the high dose range.

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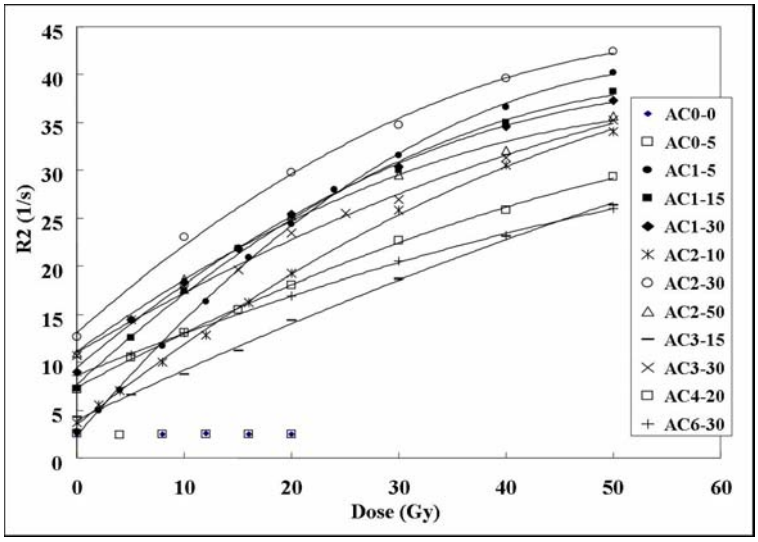
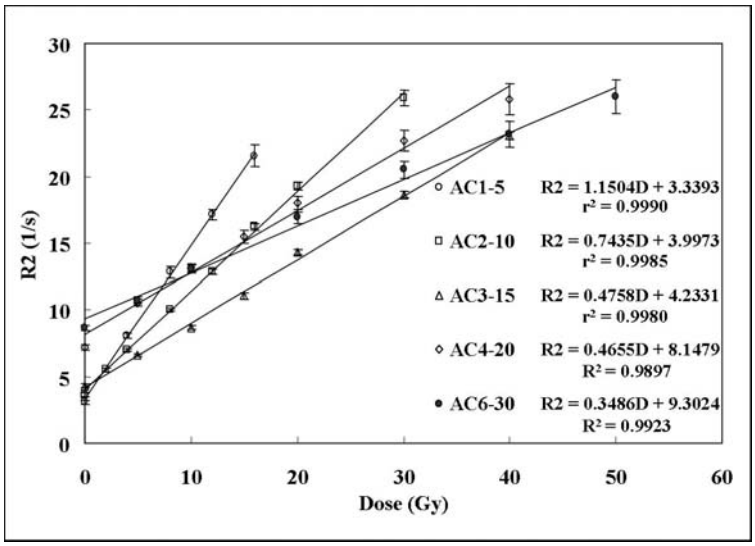


(a)



(b)

Figure 1. (a) The R2-dose response curves depending on ascorbic acid and copper sulfate concentration with a dose range of 0-50 Gy. (b) The linear dose response region of the AC1-5, AC2-10, AC3-15, AC4-20 and AC 6-30 gels.

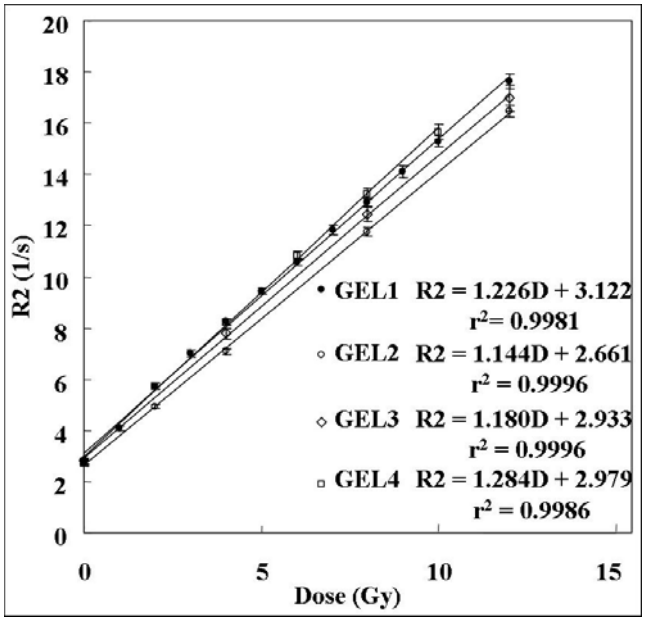
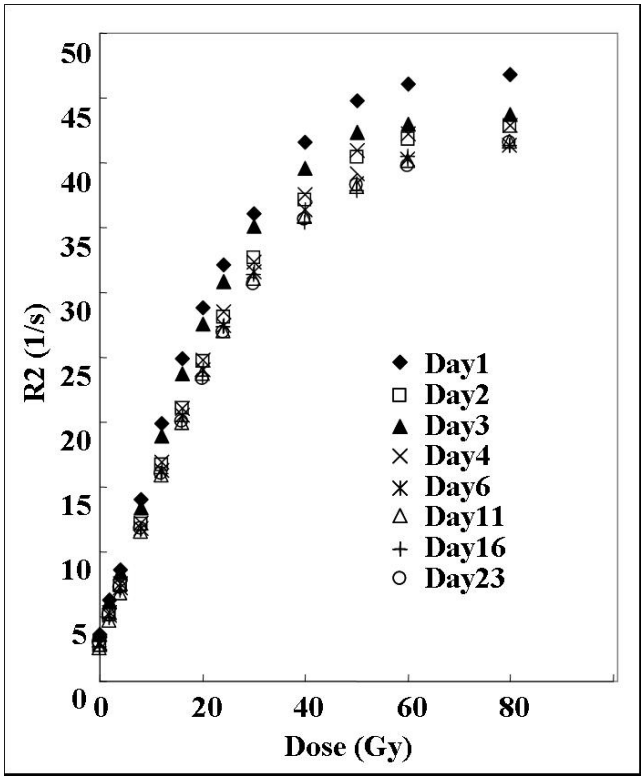
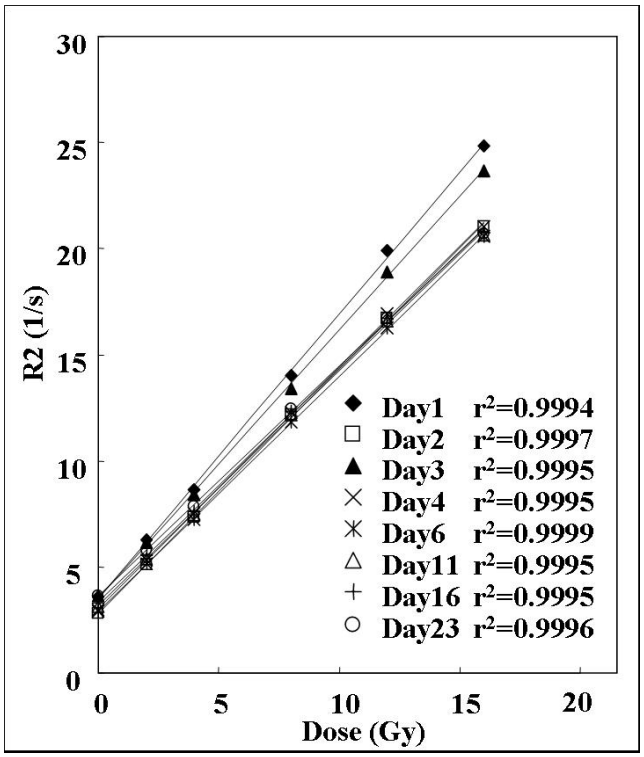


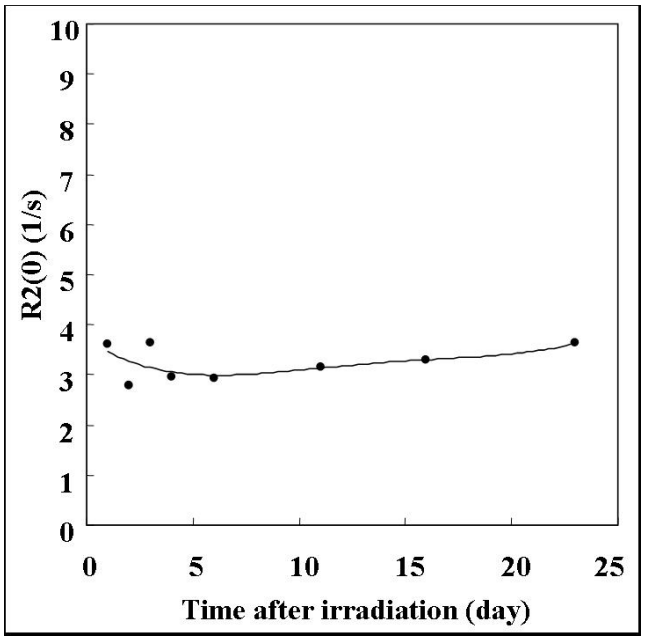
Figure 2. The R2-dose response plots measured in four separately prepared AC1-5 gels. D=dose in Grays.



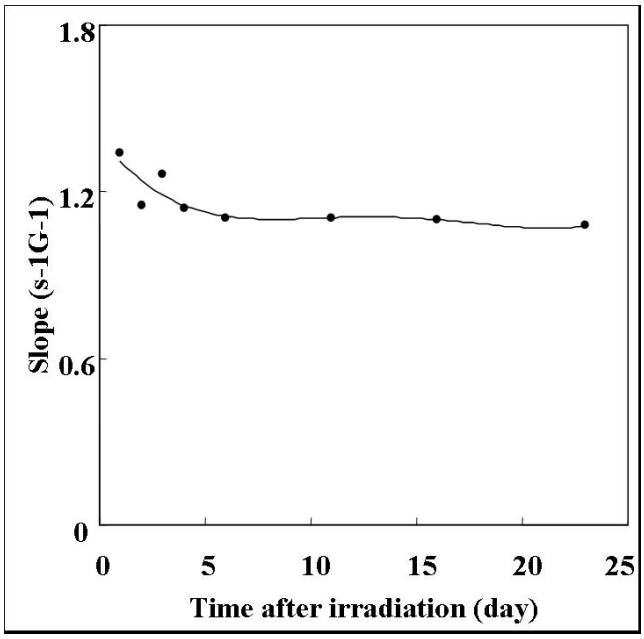
(a)


R2 (1/s)
Dose (Gy)
Day1 r²=0.9994
Day2 r²=0.9997
Day3 r²=0.9995
Day4 r²=0.9995
Day6 r²=0.9999
Day11 r²=0.9995
Day16 r²=0.9995
Day23 r²=0.9996

(b)

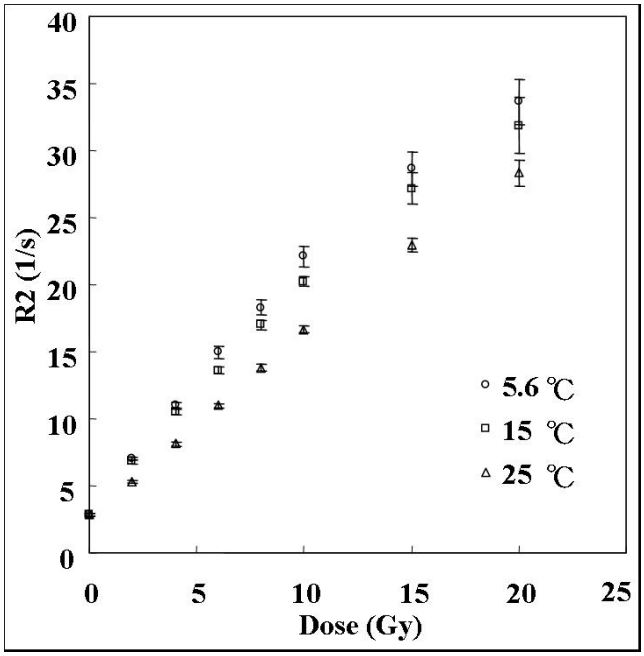

R2(0) (1/s)
Time after irradiation (day)

(c)

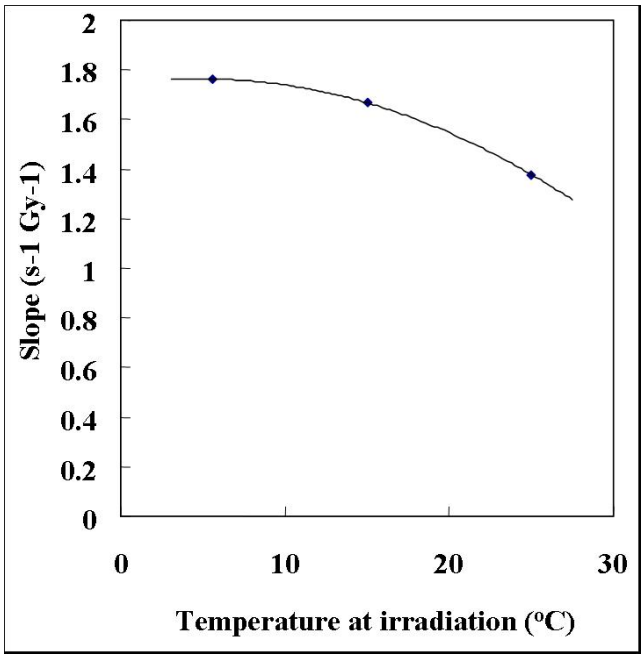


(d)

Figure 3. (a) The R2-dose response plots with times after irradiation. (b) The linear dose response regions (0-16 Gy) from Figure 3(a). (c) R2(0) values with times after irradiation. (d) The slope of the linear dose response curve up to 16Gy with increasing time.

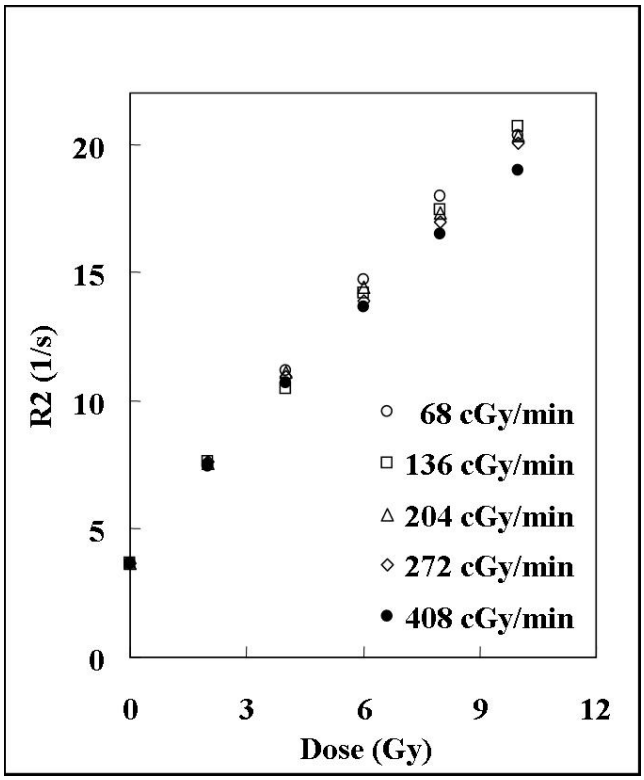


(a)

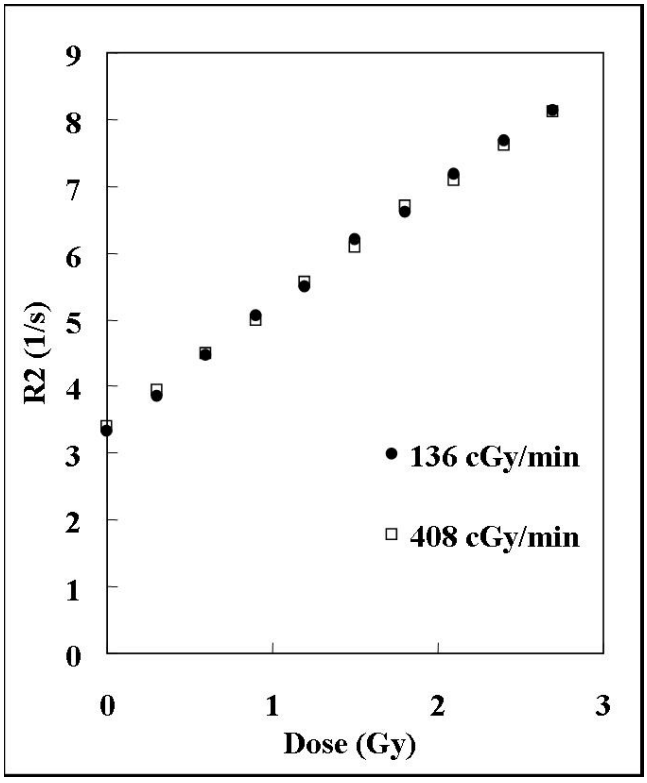


(b)

Figure 4. (a) The R2-dose response plots at different temperatures at the time of irradiation. (b) The dependence of the slope on the temperatures at the time of irradiation.



(a)



(b)

Figure 5. (a) The R2-dose response plots at various dose rates. (b) The R2-dose response plots at dose rate of 136 cGy/min and 408 cGy/min with a dose range of 0-2.7 Gy.