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# ***Dosimetry for radiation processing***

*Final report of the Co-ordinated Research Project on  
Characterization and Evaluation of High Dose Dosimetry Techniques for  
Quality Assurance in Radiation Processing*



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## FOREWORD

The use of large cobalt-60 gamma ray facilities and electron beam accelerators with beam energies from about 0.1 to 10 MeV for industrial processing continues to increase. Quality assurance is vital for the success of this technology; and the key element in QA in radiation processing is a well characterized, reliable dosimetry system that is traceable to the international measurement system. This is especially the case for health regulated processes, such as the radiation sterilization of health care products and the irradiation of food to eliminate pathogenic organisms or to control insect pests.

Many dosimetry systems are available and are being successfully used. However, it has been a growing concern that the impact of various influence quantities on the performance of these dosimetry systems is not satisfactorily understood. Besides, there are the prospects of continued expansion of the radiation processing industry through the use of new high power, high energy electron accelerators to produce intense electron beams or highly penetrating X rays (bremsstrahlung). Hence, there are new challenges for achieving harmonization in quality control in production as well as pilot-scale environments.

To meet some of these challenges, including improving current dosimetry techniques and procedures, a Co-ordinated Research Project (CRP) on Characterization and Evaluation of High Dose Dosimetry Techniques for Quality Assurance in Radiation Processing was initiated by the IAEA in June 1995. The major goals of this CRP were to investigate the parameters that influence the response of dosimeters and to develop reference and transfer dosimetry techniques, especially for electron beams of energy less than 4 MeV and for high energy X ray sources (up to 5 MV). These should help unify the radiation measurements performed by different radiation processing facilities and other high dose dosimetry users in Member States, and encourage efforts to obtain traceability to primary and secondary standards laboratories. It should also help strengthen and expand the present International Dose Assurance Service (IDAS) provided by the IAEA.

During the CRP, there were two research co-ordination meetings where the progress of the individual participants was reviewed together with the progress of the CRP as a whole. Near the end of the CRP (May 1999), a consultants' meeting was convened by the IAEA to help prepare the final report for the CRP describing the results and conclusion of the research carried out by the participants of the CRP. Prior to this meeting all the participants had submitted their individual final reports; these were reviewed by the consultants before preparing the CRP final report. The consultants were R. Chu (Canada), A. Miller (Denmark) and A. Kovács (Hungary). T. Kojima also participated at no cost to the IAEA. The draft was then circulated to all the CRP participants for their comments and suggestions before finalising the report.

This final report is published here as an IAEA TECDOC since it is realized that the contributions of this CRP are invaluable to both the operators and users of production irradiators and to those involved in the supply, calibration and performance of dosimetry systems.

The staff member responsible for the preparation of this document was K. Mehta from the Division of Human Health, who was the scientific secretary of the CRP.

### *EDITORIAL NOTE*

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## 1. INTRODUCTION

In many Member States the use of large cobalt-60 gamma ray facilities and electron beam accelerators with beam energies from about 0.1 to 10 MeV for industrial processing continues to increase. For these processes, quality assurance relies on the application of well established dosimetry systems and procedures. This is especially the case for health regulated processes, such as the radiation sterilization of health care products, and the irradiation of food to eliminate pathogenic organisms or to control insect pests.

For radiation sterilization, the publication and implementation of the international standard ISO 11137 — Sterilization of Health Care Products — Requirements for Validation and Routine Control — Radiation Sterilization and the European Standard EN 552 — Sterilization of Medical Devices — Validation and Routine Control of Sterilization by Irradiation, have resulted in the standardization of requirements to ensure global harmonization. These documents refer to dosimetry as a key element for validation of a process. Standardized dosimetry is also valuable in the radiation processing of many widely used commodities, such as polymers, automotive and airborne components, battery parts, computers, audio and video hardware, coatings, lubricants, adhesives, and composites.

The growing worldwide interest in the use of radiation processing to improve the environment has stimulated research in the large scale treatment of many solid, fluid, and gaseous wastes, bringing new requirements for quality control by dosimetry and process parameter monitoring. There are also the prospects of continued expansion of the radiation processing industry through the use of new high power, high energy electron accelerators to produce intense electron beams or highly penetrating X rays (bremsstrahlung). Hence, there are new challenges for achieving harmonization in quality control in production as well as pilot-scale environments. Also, more reliable methods for dose measurements would encourage the industry to choose suitable radiation technology over a classical approach (for example, sterilization by radiation as against EtO).

A co-ordinated research project (CRP) was initiated by the IAEA in June 1995. Research contracts and research agreements in areas of high dose dosimetry were initiated to meet these challenges. The major goals of this CRP were to investigate the parameters that influence the response of dosimeters and to develop reference and transfer dosimetry techniques, especially for electron beams of energy less than 4 MeV and for high energy X ray sources (up to 5 MV). These will help to unify the radiation measurements performed by different radiation processing facilities and other high dose dosimetry users in Member States and encourage efforts to obtain traceability to primary and secondary standards laboratories. It will also aim to strengthen and expand the present International Dose Assurance Service (IDAS) provided by the IAEA.

## 2. WORK PERFORMED

This section gives the specific objectives proposed to allow comparisons between the progress achieved and the stated goals of the CRP. It also describes the work performed to meet each of these objectives; highlights from the final reports submitted by the participants are given here to show how these objectives were addressed. The name of the CRP participant who has contributed to the particular research is shown in the parenthesis in the text.

The full comprehensive reports from each participant are given in the Annex. For a CRP of this nature, much activity is centred around research and development. This is reflected in several papers being published by the participants in scientific journals reporting the research carried out under the scope of this CRP. This amounts to 31 papers published during the time span of the CRP; these are listed in Section 3.

To ensure consistency between measurements performed by the different participants, an intercomparison was performed near the beginning of the CRP to test the ability of the participants to deliver a specified dose. Alanine dosimeters from the IAEA were distributed to the participants for irradiation in their cobalt-60 irradiators and analysis was performed by the IAEA using the IDAS procedure. Further details of the intercomparison are given in Appendix I.

## **2.1. First objective**

*The first objective was to understand and evaluate the influence of various external parameters on the performance of routine dosimeters in use at the present. Such parameters could include irradiation temperature, humidity, radiation energy, radiation type, oxygen content, light, and dose rate. It was important to study the effect of more than one parameter simultaneously. This should help increase the accuracy and reliability of the routine dosimetry systems.*

To meet this objective, it is first necessary to understand the influence of individual analysis parameters and the effect of influence quantities on the performance of the present dosimetry systems.

### **2.1.1. Analysis parameters**

For some dosimetry systems, such as those requiring evaluation by optical spectrophotometry and EPR analysis, there are a number of analysis parameters that may be adjusted to obtain optimum results. Since EPR spectrometry has more variables than other methods, studies on the optimization of the analysis parameters for EPR spectrometer were performed. A multi-parametric approach (experimental design) was used to obtain the best signal/noise ratio with the highest amplitude of the central line of the EPR signal, and the best reproducibility and resolution. Thermal sensitivity of the EPR spectrometer diodes was found to be significant in EPR spectrometry (Dolo).

The effects of analysis temperature were determined for dichromate and alanine. For EPR analysis of alanine at temperatures between 0–50°C, the temperature coefficient was found to be –0.25%/°C over the dose range from 0.1–10 kGy. This temperature effect is generally compensated by frequent measurement of reference samples. The effect of the analysis temperature for both the low dose and high dose silver dichromate dosimeters was found to be not appreciable (Kojima). Similarly, the orientation and positioning of the alanine dosimeter during analysis are critical and the influences on the uncertainty of the results were analyzed for different shapes of alanine dosimeters (rods and thin films) and for dosimeters prepared by different moulding procedures (Dolo, Kojima, Zagorski).



## **2.1.2. Influence quantities**

### *2.1.2.1. Temperature*

During the gamma sterilization of medical devices in commercial production facilities, the product temperature may increase by as much as 40°C. This temperature increase is the result of absorption of gamma energy in product and surrounding materials in the irradiation room and may have seasonal variations. For electron irradiation, most of the dose is received in a short time so the temperature increase is mostly due to the absorption of the electrons. For the irradiation of refrigerated foods, the temperature is controlled to ensure that the product temperature is between 0 and 8°C. For frozen foods, the temperature is maintained at about -20°C. Other applications may require irradiations in dry ice at -78°C, e.g. the sterilization of tissues, or in liquid nitrogen at -196°C. When the dependence of the dosimeter response on temperature is linear, the dependence can be described by a temperature coefficient as a percentage change (either in response or in dose) per °C.

Measurements of temperature rise in products were performed to determine the temperature experienced by routine dosimeters at different times during the irradiation cycle. The complexity of the temperature profile in industrial gamma facilities was demonstrated by on-line measurements using a thermocouple with a transmitter and with a bimetallic recording thermometer. For dosimeters with known temperature coefficients, the weighted average irradiation temperature can then be used to correct the dosimeter response for temperature. Another approach, which is more suitable for transfer dosimetry, is to use pairs of dosimeters with negative and positive temperature coefficients of similar magnitude to obtain a temperature independent dose determination. This approach was tested with ceric-cerous and alanine dosimeters in a gammacell and at a commercial production irradiator. Measurements with pairs of ceric-cerous and alanine dosimeters demonstrated that the average value of the doses measured by the dosimeter-pair calibrated for an arbitrary irradiation temperature was in good agreement with the dose obtained for one of the dosimetry systems with a correction applied for the effective irradiation temperature estimated from the ambient and maximum temperatures. Alternatively, the results could be used to obtain an estimate of the effective irradiation temperature (Chu).

The temperature effect was studied for several reference dosimeters (alanine, ECB, glutamine) as well as for many routine dosimeters. Different dose ranges and the temperature ranges were selected for the different dosimetry systems, depending on the applicability of each system.

To extend the applicability of alanine-polystyrene dosimeters, the effect of the irradiation temperature on the response characteristics was studied for lower irradiation temperatures down to -196°C at an absorbed dose of 5 kGy. The temperature coefficient in the range from -78 to 70°C is 0.24% of the response/°C, while a larger coefficient was indicated at -196°C (Kojima). For the NIST alanine-polyethylene dosimeters, studies were performed for irradiation temperatures from 10 to 50°C at absorbed doses of 3 to 100 kGy. The temperature coefficient ranged from 0.13% of the response/°C at 20 kGy to 0.19% of the response/°C at 100 kGy (McLaughlin).

The effect of the irradiation temperature on ethanol chlorobenzene (ECB) dosimeters was studied for temperatures from -30 to +50°C and the temperature coefficient was found to be approximately 0.06% of the dose/°C for doses of 1, 10 and 30 kGy (Kovács). The effect of the irradiation temperature on glutamine dosimeters was studied for temperatures from 15 to 55°C

for the spectrophotometric response. The response in the dose range from 5 to 40 kGy has a complex dependence, resulting in variations in the order of  $\pm 10\%$  (Gupta). The temperature coefficient for silver dichromate was re-evaluated in the temperature range 25 to 50°C, and was found to be  $-0.20$  and  $-0.26\%$  of the response/°C for doses 2–10 kGy and 10–50 kGy, respectively (Kojima).

The effects of the irradiation temperature on dose responses of several commercial routine dosimeters (Harwell Red 4034, Amber 3042, Gammachrome YR, Radix RN-15, FWT-60-00, FWT-70-40m, GafChromic DM-1260 and CTA FTR-125) were studied at dose rates ranging from 0.2 to 4 Gy/s over the temperature range  $-196$  to 60°C. The results indicate that these effects cannot be represented by constant temperature coefficients or simple relationships between response and temperature. Further studies are needed under practical irradiation conditions that are similar to those in the plant (Biramontri, Kojima).

#### *2.1.2.2. Dose rate*

In production irradiators, dose rates vary significantly depending on position of the dosimeter relative to the source. Dose rates can vary from less than 1 kGy/h to over 100 kGy/h in irradiators containing megacurie quantities of cobalt-60 and also depend on irradiator design.

Real-time measurements using an ionization chamber and a radio transmitter were performed at several irradiators to determine the dose rates to which product were subjected during processing in production irradiation facilities. These data were consistent with dose rates estimated by mathematical modelling performed using point kernel calculations. These data provide a clear indication of the magnitude of the dose rate variation and emphasize the need to have dosimeters which are not significantly affected by dose rate and for an in-plant calibration (Chu).

Response functions for specific batches of several commercial routine dosimeters (Harwell Red 4034 and Gammachrome YR, FWT-60-00 radiochromic films, FWT-70-40 optical waveguides, GafChromic films, and Fuji CTA-FTR-125 films) were determined at dose rates between 0.2 and 4 Gy/s. For FWT-70-40 optical waveguides and Gammachrome YR at doses up to 1 kGy, only slight variation in the response of the dosimeter was found for different gamma dose rates in the range from 0.2 to 4 Gy/s for irradiation temperatures from  $-18^\circ\text{C}$  to  $35^\circ\text{C}$ . Concerning CTA-FTR-125 films, significant dose rate effects were found at 25 and  $35^\circ\text{C}$  irradiation temperatures. For GafChromic DM-1260 dosimeter media, some dose rate effects were found at doses above 1 kGy. For FWT-60-00 batch 7F7 film dosimeters, higher responses were found at higher dose rates (Biramontri).

#### *2.1.3. Conditions after irradiation*

Post-irradiation conditions such as temperature and humidity affect the stability of the response of several dosimeters. Also, these effects may be different at different dose levels. Short term effects, minutes to days, are a concern for some dosimeters such as alanine and glutamine. Long term effects, days to weeks, are a concern for several other systems.

To understand the combined effects of multiple variables, a number of experiments are required. However this number may be minimized through using the method of experimental design. Preliminary studies using this technique were carried out to understand the time behaviour of alanine response after irradiation (Dolo).

### *2.1.3.1. Short term stability*

Studies at NIST with alanine-polyethylene dosimeters showed that at relative humidity up to 60% the signal was stable to within  $\pm 1\%$  up to several hours after irradiation. However at higher relative humidity, the signal increases during the first few hours (McLaughlin). Studies at BARC on the stability of glutamine solution indicated that fresh FX solution should be prepared and used within a few hours. After dissolution, the solution should be kept for about 30 minutes before analyses for the reactions to complete (Gupta).

Studies of short term post-irradiation stability were performed for Harwell Red 4034 and Gammachrome YR, FWT-60-00, OptiChromic, GafChromic, and CTA-FTR-125. For Harwell dosimeters and CTA-FTR-125 only slight variation in response was found for the period of 1 hour to 7 days, while no variation in response was found for GafChromic, FWT-60-00 and FWT-70-40 (Biramontri).

### *2.1.3.2. Long term stability*

Such studies were performed for alanine (McLaughlin, Dolo) and glutamine (Gupta). Studies at NIST showed that for alanine-polyethylene dosimeters the higher the dose and the higher the relative humidity, the greater the fading rate. The EPR signal decreased by only about 0.3% for the relative humidity of 33–44% for 10 kGy (McLaughlin). The post-irradiation stability of glutamine dosimeter was found to depend on dose and temperature of storage. For example, the decrease in response was negligible for 4 months at 25°C for 20 kGy. However, it was about 3.6% for 60 kGy (Gupta).

### **2.1.4. Other studies**

Understanding the kinetics of the chemical reactions can lead to improved performance relative to influence quantities. One of the methods for accomplishing this, for example, is through studying the short as well as long term evolution of the dosimeter response. There may be different objectives: to determine a zone of stability of measurement (no apparent evolution of the kinetics), or to identify the important parameters that influence the dosimeter response. This then can give a quantitative correction coefficient with the associated limits of its application, even if it is just a mathematical approach without coherence with physics or chemistry. For such study, a qualitative and quantitative model is needed to describe the mechanism that is the basis of the dosimetry technique. Such studies were performed for alanine dosimeters with the method of experimental design.

## **2.2. Second objective**

*The second objective was to develop reference, transfer and routine dosimetry techniques, especially for electron beams of energy less than 4 MeV. This would facilitate extension of the present IAEA 's dose assurance service (IDAS) to these radiation types.*

To meet this objective, some existing dosimetry systems were modified and new reference and routine dosimeters suitable for electron beams of energy up to 4 MeV were developed and characterized.

### **2.2.1. Modifications of existing systems**

Some existing dosimeters were modified to be applicable for low energy electrons.

Thin polystyrene calorimeters suitable for electron energies of 1.5 MeV or higher were developed and tested. Procedures for the calibration of these calorimeters were established, as well as methods for their use for calibration of other routine dosimeters. The thermal stability of the calorimeters for use at 1–4 MeV electron accelerators was studied. Because of the heat losses from the calorimeter after irradiation, a procedure for its use must be established for each specific accelerator facility (Miller).

Alanine film analysis based on diffuse reflectance spectrometry (DRS) was shown to have promise to provide an alternate to EPR spectrometry. Advantages include the lack of dosimeter orientation effect during analysis, no dependence on alanine mass since the measurement occurs at the surface, simple analysis procedure, and less expensive equipment (Zagorski).

Glutamine powder in 0.1 mm thick packages was studied for electron beam dosimetry from 0.4 to 10 MeV, and was successfully used for determining the depth dose distribution for a 2 MeV electron beam (Gupta). Applicability of ECB solution in 5 mm thick double layer bags was investigated, and was found to be suitable for 4 MeV electrons (Kovács).

### **2.2.2. New film dosimeters**

Film dosimeters suitable for low energy electron irradiation were developed and tested.

SUNNA dosimeters were characterized for both gamma and electron irradiations for doses from 10 Gy to 5 kGy based on optically stimulated luminescence (OSL) and for doses from 5 kGy to 100 kGy based on spectrophotometric evaluation at 240 nm. The response functions were found to be significantly different for gamma rays and electrons. The effect of irradiation temperature from 15 to 50°C was studied, and the temperature coefficient was found to be less than 0.1% of the dose/°C in the dose range 10 to 30 kGy (spectrophotometric evaluation). No effect of irradiation temperature was observed for doses below 1 kGy, while significant temperature effects occur above this level (OSL evaluation). For spectrophotometric evaluation, the readings were stable for 2 to 3 days following irradiation and fading was observed for longer periods. A heat treatment (60°C for 15 minutes) was found useful to stabilize the OSL signal for a period of several weeks (Kovács).

PVG film dosimetry based on spectrophotometric analysis was found to be suitable for gamma irradiations for doses from 5 to 100 kGy and for electron beam irradiations from 5 to 140 kGy. The response functions were found to be somewhat different for these two radiation qualities. The effect of irradiation temperature from 20 to 55°C was studied for electrons, and the temperature coefficient was found to be 0.8% of the response/°C in the dose range from 4 to 55 kGy. Irradiated PVG film was stable at room temperature with a variation within  $\pm 3\%$  over a storage period of 2 months. Studies on colour development showed that dosimeters should be stored for 24 h after irradiation before they are analysed (Chen).

## **2.3. Third objective**

*The third objective was to investigate the suitability of using existing dosimetry systems for X rays.*

Application of X rays for radiation processing is promising in the near future as an alternative radiation source to  $^{60}\text{Co}$ . Before existing dosimetry systems which have been well characterized for  $^{60}\text{Co}$  gamma rays and electron beams can be used for this radiation quality, their performance under X rays should be evaluated.

Behaviour of undyed PMMA dosimeter (Radix) and alanine-polystyrene rod dosimeter was studied for X rays obtained from 3 MeV electrons (current = 20 mA) over the dose rate range from 5 to 400 kGy/h. Temperature rise during irradiation was monitored and was found to be insignificant. Both dosimeters showed negligible dose rate dependence over this range (Kojima).

Also, a dose intercomparison study was conducted among several dosimetry systems for X rays obtained from a 5 MeV electron accelerator. All the dosimeters employed in this study were well characterized for  $^{60}\text{Co}$  gamma rays at each participating laboratory. Also, almost all of them have been used for electrons. There were nine types of dosimeters provided by the participants, and three dosimeters of each type were irradiated at each of the three dose levels: 4, 8 and 12 kGy. Additional details of irradiation conditions are given in Appendix II.

The electron accelerator, a Dynamitron (5 MeV, 10 mA, scan width: 96 cm) operated by West Japan Irradiation Service Co., Ltd, Osaka, used a tantalum target to provide X rays. A conveyor speed of 2.0 m/min was used for this study which yielded a dose rate of about 0.2 kGy/pass. Several polystyrene phantoms were used to accommodate the dosimeters, and 20 mm thick PMMA plates were placed above and below them to avoid contamination of scattered electrons, to establish secondary electron equilibrium conditions, and to stop backscattered electrons from the steel tray.

At 4, 8, and 12 kGy levels, the mean and the standard deviations of the estimated values for all the dosimetry systems were  $4.17 \pm 0.09$ ,  $8.27 \pm 0.22$ , and  $12.38 \pm 0.24$  kGy, respectively. The average coefficient of variation of all the 27 measurement points is about 2.1%. Considering the influence of various parameters such as energy spectrum, dose rate distribution in lateral direction and along the beam axis, irradiation temperature, and humidity, the agreement of 2.1% among the various dosimeters was within the reasonably achievable limit for high dose dosimetry. More details are given in Appendix II. The results were similar to the previous dose intercomparison study for  $^{60}\text{Co}$  gamma rays, although in that case only one type of dosimeter (alanine dosimeters of IAEA) was irradiated at several participating laboratories (see Appendix I).

The above results suggest that all the dosimetry systems used in this study, which are calibrated for  $^{60}\text{Co}$  gamma rays, have a potential to be used for X rays produced by high energy electron beams.

To further understand the behaviour of PMMA and alanine dosimeters, their effective mass energy absorption coefficients were calculated for  $^{60}\text{Co}$  gamma rays and for X ray spectra from 3, 5 and 7 MeV electrons (Kojima).

#### **2.4. Fourth objective**

*The fourth objective was to establish suitable irradiation geometries to achieve consistency in dose calibration and interlaboratory comparison.*

Standard irradiation geometries (phantoms) for gamma rays, electrons and X rays were proposed for minimizing variations in doses received by different dosimeters irradiated together. These phantoms were used for the X ray intercomparison (see Appendix II).

For intercomparison and in-plant calibration purposes, it is necessary to irradiate several dosimeters to the same dose. To avoid dose variations caused by the irradiation geometry, it is recommended that the dosimeters be irradiated together in a standard absorber or a phantom. The phantom material should be similar to the material of the dosimeter. Due to the different nature of gamma and electron radiation, phantoms of different design are needed.

For electron beam irradiations, the material thickness depends on the electron energy. Appropriate amount of material should be placed in front of the dosimeter so that it is completely in the increasing linear portion of the depth dose curve. Also, sufficient material should be placed behind the dosimeter to provide proper backscattering of electrons. This phantom is generally irradiated separately from the product.

Similar considerations apply for gamma and X ray irradiations. The dosimeters should be surrounded by material similar to the dosimeter, e.g. polystyrene, in order to decrease the effect of interfaces. There should be sufficient amount of material on all sides to achieve the conditions of electron equilibrium. The phantom should be placed in a region of low dose variation, e.g. in the middle of a homogeneous dummy product.

Additional details of the suggested phantoms are given in Appendix III.

### 3. SCIENTIFIC PUBLICATIONS

These are articles published in refereed scientific journals and other publications by the chief scientific investigator or their staff describing work related to this CRP. Those directly connected with the research contract or agreement with the IAEA are identified by an asterisk (\*).

#### 1995

1. KOJIMA, T., KASHIWAZAKI, S., TACHIBANA, H., TANAKA, R., DESROSIERS, M.F., McLAUGHLIN, W.L., Orientation effects on ESR analysis of alanine-polymer dosimeters, *Appl. Radiat. Isot.* **46** (1995)1407–1411.
2. MILLER, A., Polystyrene calorimeter for electron beam dose measurements, *Radiat. Phys. Chem.* **46** (1995) 1240.
- 3.\* ZAGÓRSKI, Z.P., RAFALSKI A., A thin alanine-polyethylene film dosimetry system with diffuse reflection spectrophotometric evaluation, *J. Radioanal. Nucl. Chem. Articles* **196** (1995) 97–105.

#### 1996

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- 7.\* LIU, A., CHEN, W., Radiation effect on solution LMG acidic aqueous and ethanol, Proceedings of 7th China–Japan Bilateral Symposium on Radiation Chemistry, Oct. 28–Nov.1, 1996, Chengdu, China (1996) 58–163.
- 8.\* PRZYBYTNIAK, G.K., ZAGÓRSKI Z.P., Orientation of crystals in alanine dosimeter assessed by DRS, as seen in EPR spectra evaluation, *J. Radioanal. Nucl. Chem. Letters* **212** (1996) 373–382.

#### 1997

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10. KOJIMA, T., KASHIWAZAKI, S., ZHANG, Y., Alanine-polystyrene dosimeters prepared by injection moulding. *Appl. Radiat. Isot.* **48** (1997) 965–968.
11. ZAGÓRSKI, Z.P., PRZYBYTNIAK, G.K., Optical properties of the radical anion derived from  $\alpha$ -alanine by deamination. *Nukleonika* **42** (1997) 373–378.

#### 1998

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## 4. CONCLUSIONS

Research conducted by the participants under the CRP provided valuable contributions adding to the understanding of both the unique dosimetry requirements of the radiation processing industry and the characteristics of the currently used dosimetry systems. The main results, in relation to the four objectives, are summarized below:

*Objective 1:* Information gathered from the studies performed by the participants improved the knowledge of the influence of various analysis parameters and influence quantities on the response of dosimeters. These parameters include dosimeter positioning during analysis, dosimeter temperature and humidity before, during and after irradiation, dose rate and post-irradiation stability. This information helps to increase the accuracy and the reliability of the dosimetry results and allows better understanding of the results of dose intercomparisons and dose verification, e.g. IDAS. At the same time it highlighted the difficulties in the interpretation of the dosimetry results when the dosimeter response is affected by more than one parameter. This confirms the need to verify the applicability of the calibration relationship under processing conditions.

*Objective 2:* The usefulness of a new thin polystyrene calorimeter was demonstrated for the routine monitoring for electron beams of low energies (as low as 1.5 MeV) and for the calibration of routine film dosimeters and alanine pellets. Modifications of present dosimeters demonstrated that powder and liquid dosimeters used for higher electron energies may also be used for lower energies when packed in appropriately thin packages. New routine film dosimeters show promise for use for low energy electron beams.

*Objective 3:* The X ray intercomparison results suggest that all the dosimetry systems used in this study (namely alanine, dichromate, ceric–cerous, ECB, glutamine) have a potential to be used for X rays produced by high-energy electron beams, when calibrated using  $^{60}\text{Co}$  gamma rays.

*Objective 4:* New standard dosimeter holders may be useful for reducing uncertainties in in-plant calibrations, verifications and intercomparisons through better control of the irradiation conditions.

As shown above, the contributions of the CRP are invaluable to both the operators and users of production irradiators and to those involved in the supply, calibration and performance of dosimetry systems. The contributions of the IAEA in this area have been, and continue to be a vital source of leadership, resulting in improved processes and products. This information should be readily made available to the radiation processing industry.

Studies such as those performed under the CRP are worthwhile and warrant continuation to ensure that the science of dosimetry continues to progress. One area to be emphasized in dosimetry research is that the observed qualitative results should be interpreted in terms of radiation chemical reaction mechanisms. For example, better understanding is required of the origin of the high temperature-coefficient of the free radical formation in irradiated alanine.



## Appendix I

### COBALT-60 GAMMA RAYS INTERCOMPARISON

#### Introduction

An intercomparison in  $^{60}\text{Co}$  gamma fields, using IDAS alanine dosimeters, was performed to ensure that all doses quoted by the CRP participants are in agreement. This 'double-blind' intercomparison was carried out in the summer of 1997.

#### Protocol

The protocol that was followed was similar to the one developed by the BIPM earlier for the 1995 intercomparison for the high-dose calibration laboratories [1]. The protocol used for this intercomparison is reproduced below.

*Issuing laboratory:* IAEA Dosimetry Laboratory

Participating irradiating laboratories: All 10 participants of the CRP

*Radiation source:*  $^{60}\text{Co}$

*Gamma dose range:*  $15 \pm 3$  kGy (dose to water)

*Issuing laboratory to provide* for each irradiating laboratory four alanine dosimeters in polystyrene capsules, along with instructions for pre- and post-irradiation care.

*Dosimeter dimension:* The polystyrene capsule is 50 mm long and 12 mm diameter. The wall thickness is 4 mm; enough to provide electron equilibrium for  $^{60}\text{Co}$  gamma rays. The alanine dosimeter inside the capsule is 30-mm long and 3-mm in diameter.

*Irradiation:*

- Three (3) dosimeters to be irradiated to the same dose (preferably together in the black package); the exact dose level is to be chosen by each irradiating laboratory between 12 and 18 kGy.
- Because of the finite size of the dosimeter, it is important that (i) the gamma field is fairly uniform over its entire volume; and (ii) any perturbation in the gamma field caused by the dosimeter is estimated and corrected for.
- The irradiation temperature of the dosimeters shall be monitored and controlled within  $\pm 2^\circ\text{C}$ . The temperature shall not exceed  $40^\circ\text{C}$ . The temperature coefficient of our alanine dosimeters is  $+0.0023$  of the response/ $^\circ\text{C}$ .
- The 4<sup>th</sup> dosimeter is the 'control dosimeter' and shall always be kept with the others except when they are irradiated. The participating laboratory shall not irradiate this dosimeter. It will be irradiated to about 2 kGy at the IAEA laboratory before dispatching of the dosimeters.

*Issuing date:* early June 1997

*Return date:* within 5–6 weeks of the receipt of the dosimeters (to be returned by express mail). They should be in Vienna by 15 August 1997.

*The irradiating laboratory to send to:* O. Güven (IAEA)

- conditions of irradiation, including phantom material, irradiation temperature of the dosimeters, dose rate or duration of irradiation;
- description of your dosimetry system, including calibration and traceability to national standards;
- details of the derivation of the dose estimates;
- value of the absorbed dose delivered to the dosimeters;
- full statement of uncertainty — to be expressed in terms of  $1\sigma$  (standard deviation).

*The irradiating laboratory to send to the issuing laboratory:* K. Mehta (IAEA), besides the three irradiated dosimeters and the control dosimeter, the values of the following critical parameters:

- measured dosimeter temperature (within  $2^\circ\text{C}$  accuracy),
- date of irradiation.

*The IAEA (Mehta) to send,* after reading of the dosimeters, the estimated dose values (including corrections) to each participating laboratory and to O Güven.

*O. Güven to then release to K. Mehta* the data received from the participants for analysis.

*K. Mehta to write* the final report to be discussed at the RCM (6–9 October 1997, Vienna)

*Publication:* All parties shall regard the data supplied to them as confidential and shall not publish it in any form without the permission of all other parties involved. The IAEA may, at its discretion, publish the results of the intercomparison, but only in a form that does not identify the individual participants.

## **Data and results**

Table I.1 gives relevant data of the IAEA's transfer dosimetry system (alanine-ESR) used for this intercomparison exercise. The results and information about some of the important parameters are given in Table I.2.

The *second* column of Table I.2 gives the mean value of the three alanine dosimeters as analysed by the IAEA Dosimetry Laboratory, where the combined uncertainty in the calculated dose is 1.7% ( $1\sigma$ ); while the *third* column shows the standard deviation for the three individual values for a dosimeter set. The *fourth* column gives the dose value and the combined uncertainty as stated by each participant; while the *fifth* column gives the non-uniformity of the gamma field over the volume occupied by the three dosimeters of the set (as stated by the participants). The *sixth* column gives the ratio (R) of the participant's value to the IAEA value.

TABLE I.1. IAEA'S TRANSFER DOSIMETRY SYSTEM: ALANINE-ESR [2]

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**Calibration Relationship** (Batch No: 9512161)

$$\text{Dose (Gy)} = 6.734 + 243.5 \cdot R + 0.5745 \cdot R^2 + 5.035 \cdot 10^{-4} \cdot R^3 + 1.270 \cdot 10^{-5} \cdot R^4$$

R = normalized Response

The regression coefficients are applicable for the dose range : ~1.5–100 kGy.

**Calibration Conditions**

- alanine dosimeters irradiated in the IAEA Gammacell 220
- dose rate = 47.16 Gy/min (97–01–01)
- dose rate was determined by dichromate transfer dosimeters from NPL
- irradiation temperature = 25°C
- dosimeters analysed 10–20 days after irradiation.

**Dosimeter Characteristics**

- pre-irradiation conditioning, 7 weeks at 29% r.h.
  - temperature coefficient = +0.23% of the response/°C
  - fading rate = 0.008%/day
  - fading correction to be applied over  $\Delta T$  days  
where,  $\Delta T$  = (day of analysis — day of irradiation — 20) days
- 

There were ten participants in the CRP and all participated in the intercomparison. However, during the exercise Laboratory 4 realised that they had a serious dosimetry problem in their laboratory and were not able to state the delivered dose with high degree of confidence; they requested to opt out of this exercise. The two laboratories, namely 2 and 5, had the ratio R much different than unity. This indicated that they had significant problem with their dosimetry system or with the procedure. Both of these laboratories were given second set of dosimeter to repeat their irradiation. These are discussed below. Thus, these two laboratories were excluded from further analyses.

The mean and the standard deviation for the seven values of R given in Table I.2 (column 6) are: mean = 0.995 and standard deviation = 1.9%. The ratio R for each participant was then normalized to this mean value, and this normalized R is plotted in Figure I.1 for the seven participants. The bars on the left give the combined uncertainty stated by the participants (column 4), and the statistical uncertainty due to the IAEA readings are shown by the bars on the right (about 0.9%). This value includes the uncertainties of temperature and fading corrections. The three dose values (as estimated by the IAEA) for the three dosimeters within a set are also shown in Fig. 1.1.

TABLE I.2. INTERCOMPARISON RESULTS

Laboratory	IAEA estimated mean dose (kGy $\pm$ 1.7%)	Std. Dev. for 3 values (%)	Participant's estim. dose (kGy $\pm$ 1 $\sigma$ )	Non-uniformity of field	Participant IAEA	Rel. Diff.* (%)	Dose rate (kGy/h)	Irrad. Temp. (°C)
1	14.99	1.69	15.0 $\pm$ 1.09%	-----	1.001	0.07	7.69	26
2	13.10	1.03	11.8 $\pm$ 3.86%	<1%	0.901	-9.92	1.07	27
3	16.16	1.33	16.0 $\pm$ 1.1%	1%	0.990	-0.99	2.5	35
4	10.00	1.56	-----	-----	-----	-----	-----	-----
5	14.31	1.15	12.9 $\pm$ 6.7%	-----	0.901	-9.85	21.8	32
6	15.26	0.40	15.32 $\pm$ 0.73%	0.3%	1.004	0.39	0.51	30
71	15.27	1.16	15.00 $\pm$ 1.07%	<1%	0.982	-1.77	10.08	29
8	12.17	1.27	12.4 $\pm$ 1.8%	<4%	1.019	1.89	4.33	27
9	15.59	1.51	15.0 $\pm$ 3.71%	-----	0.962	-3.78	9.50	25
10	13.90	1.01	14.0 $\pm$ 0.9%	2%	1.007	0.72	9.56	23
Mean :		1.20 %			0.995	-0.50		
Std. Dev. :					1.9 %	1.9		

$$* \text{ Rel. Diff (\%)} = \frac{\text{Participant's estimate} - \text{IAEA estimate}}{\text{IAEA estimate}} \times 100$$

## Analyses

As stated above, the mean and the standard deviation for the seven values of R as given in Table I.2 are: mean = 0.995 and standard deviation = 1.9%. Five out of these seven laboratories are recognized calibration laboratories; and these values for these five laboratories are: mean = 0.997 and standard deviation = 1.05%. Thus, the mean of the dose values measured by the IAEA was within 1% of the mean of the dose values stated by these five laboratories. Also, the results were reasonably distributed and no outliers seem to be present (5 out of 7 were within  $1\sigma$  from the mean, and the other two were within  $2\sigma$ ).

The standard deviations for the three dose values for each participant had a mean value of 1.20% (column 3). A part of this uncertainty was due to the statistical uncertainty of the measurements by the IAEA (0.43%, not including fading and temperature correction contribution). The difference between these two values can be explained, at least in part, by the variation (about 1%) of the dose delivered to the dosimeters of a given set due to non-uniformity of the gamma field (column 5). The results were thus consistent with the information given by the participants.

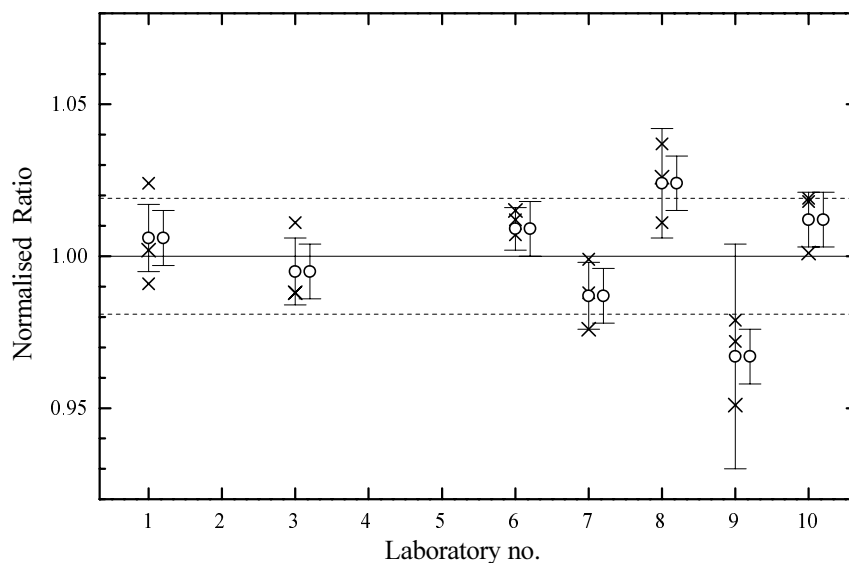


FIG. I.1. Results of the cobalt-60 gamma rays intercomparison. The ordinate is the ratio of the participant's value to the IAEA value, normalized to the mean of these ratios. The bars on the left give the combined uncertainty stated by the participants (column 4 of Table I.2), and the statistical uncertainty due to the IAEA readings are shown by the bars on the right (about 0.9%). This value includes the uncertainties of temperature and fading corrections. The three dose values (as estimated by the IAEA) for the three dosimeters within a set are also shown in the figure (x).

## **Conclusion**

The final results of the intercomparison showed that at least two laboratories were outside the acceptance limit, and possibly a third one (Laboratory 9). This pointed out the necessity of participating in such intercomparisons frequently. This also applies to commercial irradiation facilities who avoid the extra cost of the annual intercomparisons.

## **Repeat measurements**

The results from Laboratory 2 for the repeat measurements were quite different than those for the first irradiation. For the first irradiation, the distance between the dosimeters and the plane source was about 25 cm and the dose rate was 1.07 kGy/h. For the second irradiation, these values were 15 cm and 3.07 kGy/h. The Participant/IAEA dose ratio for the second irradiation was 1.024 (compared to 0.901 before!). It was later realised that a likely cause of the higher discrepancy for the first run could be that the seal of the Fricke ampoule was broken during the run, thus invalidating the Fricke measurements.

The results from Laboratory 5 for the repeat measurements were quite different also. This time the agreement with IAEA alanine dosimetry was within 2% ( $R = 1.02$ ). Completely different irradiation geometry was used for this run. The cause for the earlier discrepancy was traced to a significant non-uniformity of the radiation field due to non-uniform source loading.



## Appendix II

### X RAY INTERCOMPARISON

#### Introduction

The objective of this intercomparison was to check the performance of the existing dosimetry systems, which have been well characterized for  $^{60}\text{Co}$  gamma rays, for X rays obtained from a 5 MeV electron accelerator.

#### Experimental

##### *Dosimetry systems*

The physical and chemical properties of the nine participating dosimetry systems are listed in Table II.1. All the dosimeters are well characterized for  $^{60}\text{Co}$  gamma rays at each participating laboratory. Most of the dosimetry systems were also well characterized for electron beam radiation yielding no significant difference in response between gamma and electron irradiation. The dose rate of X ray irradiation falls typically between those of gamma and electron irradiation, and it may therefore be expected that these dosimeters are not affected by the dose rates of X ray irradiation. Three dosimeters for each type of dosimetry system were irradiated at each of the three dose levels: 4, 8 and 12 kGy. Each participant also sent at least one dosimeter as a control which was not irradiated.

TABLE II.1. DOSIMETRY SYSTEMS PARTICIPATING IN THE INTERCOMPARISON

ID Number	DOSIMETER	DIMENSIONS <sup>a</sup>
1	Alanine (30% PS)	dia = 12 mm; length = 50 mm
2	Alanine (15% PE)	dia = 13 mm; length = 30 mm
3	Alanine (15% paraffin)	dia = 25 mm; height = 6 mm
4	Alanine (30% PS)	dia = 12 mm; length = 50 mm
5	Alanine <sup>b</sup> (8% stearin) <sup>c</sup>	dia = 4.8 mm; thickness = 2.6 mm
6	Glutamine (powder)	dia = 11.8 mm; length = 32 mm
7	Dichromate (2-ml)	dia = 12 mm; length = 60 mm
8	ECB (2-ml)	dia = 11 mm; length = 80 mm
9	Ceric-cerous (2-ml)	dia = 12 mm; length = 75 mm

<sup>a</sup> This includes a holder/capsule for alanine.

<sup>b</sup> This is one pellet. A dosimeter consists of 4 such pellets; the pellets are separately placed.

<sup>c</sup> Also contains 2% of  $\text{SiO}_2$ .

##### *Irradiation condition*

The electron accelerator at West Japan Irradiation Service Co., Ltd, Osaka is a Dynamitron with the following specification: maximum energy of 5 MeV, maximum current of 10 mA, scan-horn angle of  $37.5^\circ$ , scan width of 96 cm. For the generation of X rays, a tantalum target was attached under the window at the distance of 15 cm (Fig. II.1) [3]. Variation of the beam current during irradiation was about 1%. Conveyor speed was controlled at 2.0 m/min, and its stability was checked for every pass, just before the dosimeters went below the beam window.

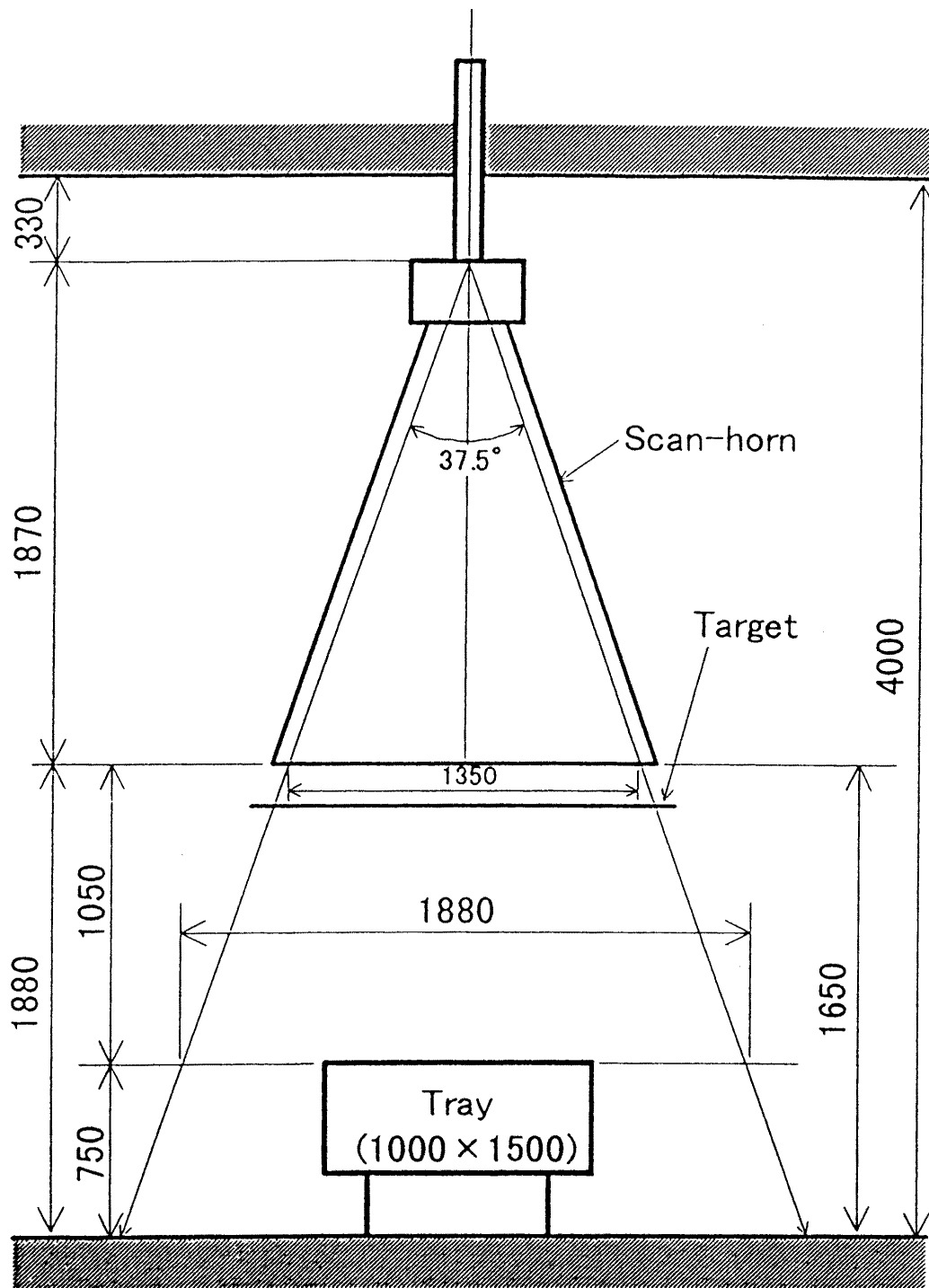


FIG. II.1. Irradiation geometry for X ray intercomparison (all dimensions are in mm).

Eighteen polystyrene flat phantoms were used to accommodate the nine types of different dosimeters to provide similar irradiation conditions. This is a disc of size 14-cm diameter and 18-mm thickness. Three dosimeters of each type were accommodated horizontally in one such phantom. Two phantoms containing the same type of dosimeters were symmetrically positioned along the center line of the scan direction for irradiation.

These phantoms were placed on two steel trays for irradiation; distance between these two trays (end-to-head) was 70 cm. The tray size was 100 cm along the scan direction and 150 cm along the conveyor-motion. Schematic diagrams of the position of these phantoms in the trays are shown in Figs II.2 and II.3.

A PMMA plate (400 mm × 400 mm, 20 mm thick) was placed above the phantoms to stop the scattered electrons from the beam reaching the dosimeters and to establish secondary electron equilibrium conditions for the X ray photons. Another similar plate was placed below the phantom for stopping the backscattered electrons from the steel tray.

Dose rate of about 200 Gy/pass at the location of the dosimeters was estimated preliminarily using FWT 60 radiochromic film dosimeters. The distribution of the dose rate was found to be better than  $\pm 2\%$  within  $\pm 200$  mm from the center of scanning. Doses of 4, 8, and 12 kGy level were given by passing dosimeters 20, 40 and 60 times under the beam, respectively. One pass took about 8 minutes, out of which the dosimeters were under the beam for 10 s.

Since dose responses of almost all dosimeters have a slight dependence on irradiation temperature, a polystyrene calorimeter with similar design to the phantoms was used to monitor the temperature after every 5 passes. The only difference is that the calorimeter core is located inside a foam box (30 cm × 30 cm), unlike the phantom with dosimeters. The measured temperature values are given in Table II.2. Ambient temperature in the irradiation room was monitored every one hour and was in the range of 16–17°C. The ambient relative humidity was in the range of 80–86% r.h. during the entire irradiation process.

TABLE II.2. IRRADIATION TEMPERATURE<sup>a</sup> OF THE DOSIMETERS

Time <sup>b</sup>	Passes	Temp. (°C)	Time	Passes	Temp. (°C)
13:40	start	15.63		after 35 passes	21.90
	after 5 passes	16.69	18:58	40	23.09
	10	17.87		45	23.46
	15	19.00		50	23.77
16:13	20	20.13		55	23.99
	25	21.09	21:30	60	24.20
	30	21.88			

<sup>a</sup> Temperature history as measured by the thermistor of the PS calorimeter that was inside a foam box (30 cm × 30 cm). Thus the geometry was similar to the irradiation phantoms, except the foam box.

<sup>b</sup> The experiment started on March 26, 1999 at 13:40 (Japanese time). 20 passes ended at about 16:13, and then it took about 20 min to remove the 4-kGy dosimeters and place the next set for 8-kGy run. The 12-kGy dosimeters were on the conveyor for the entire period. The conveyor run takes about 7 min. 50s for one full pass.

## Data and results

Table II.3 shows the results of the three runs for 4, 8, and 12 kGy levels. The third column lists the three individual dose values as well as their mean value for the three dosimeters irradiated together in one phantom. The fourth column gives the combined uncertainty ( $k=1$ ) in this mean value, as reported by each participant. The last column is the ratio of this mean value to the overall mean value of all the dosimeters. For the three dose levels, the means of the estimated values of all the dosimetry systems were  $4.17 \pm 0.09(2.1\%)$ ,  $8.27 \pm 0.22(2.7\%)$ , and  $12.38 \pm 0.24(1.9\%)$  kGy.

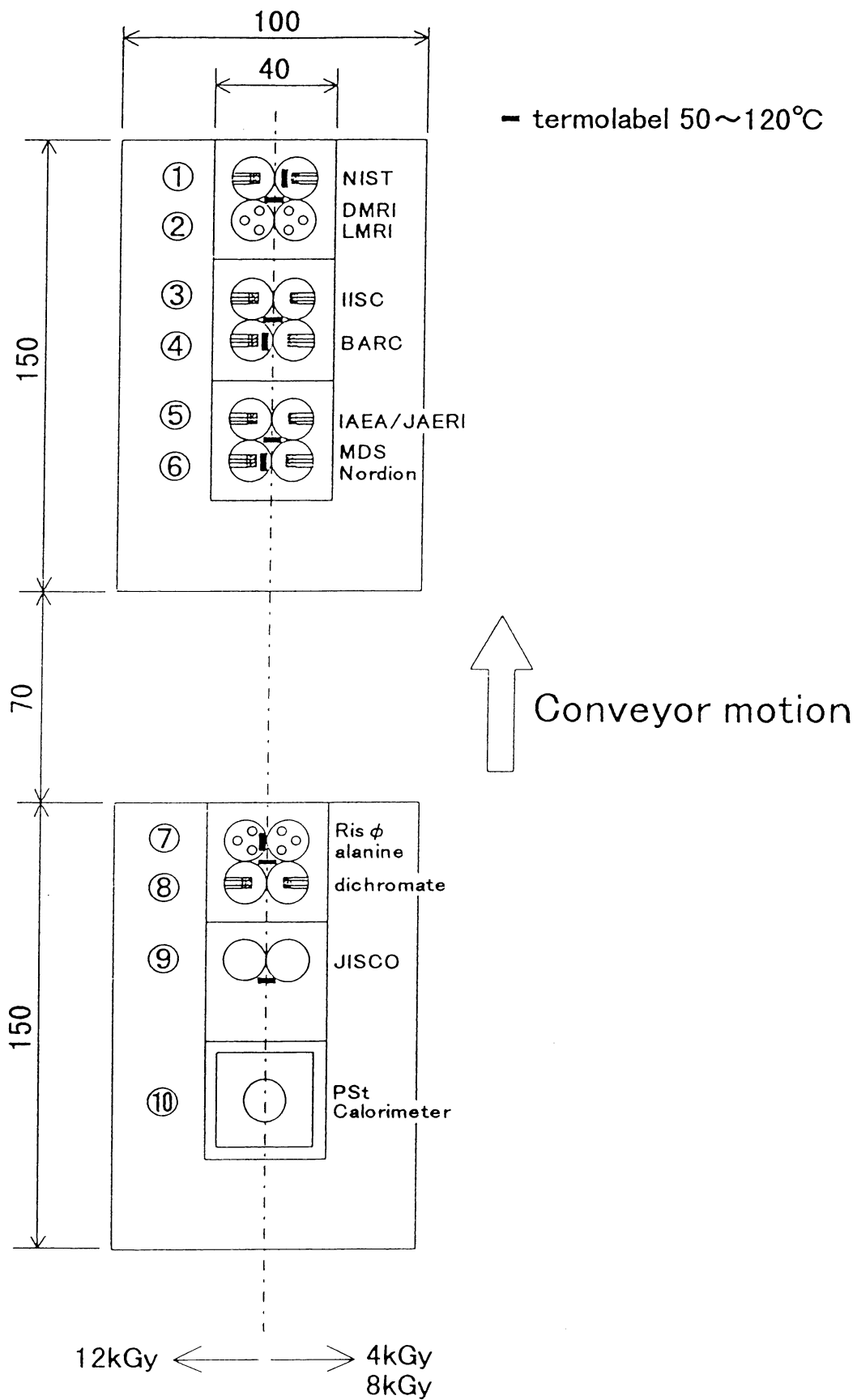
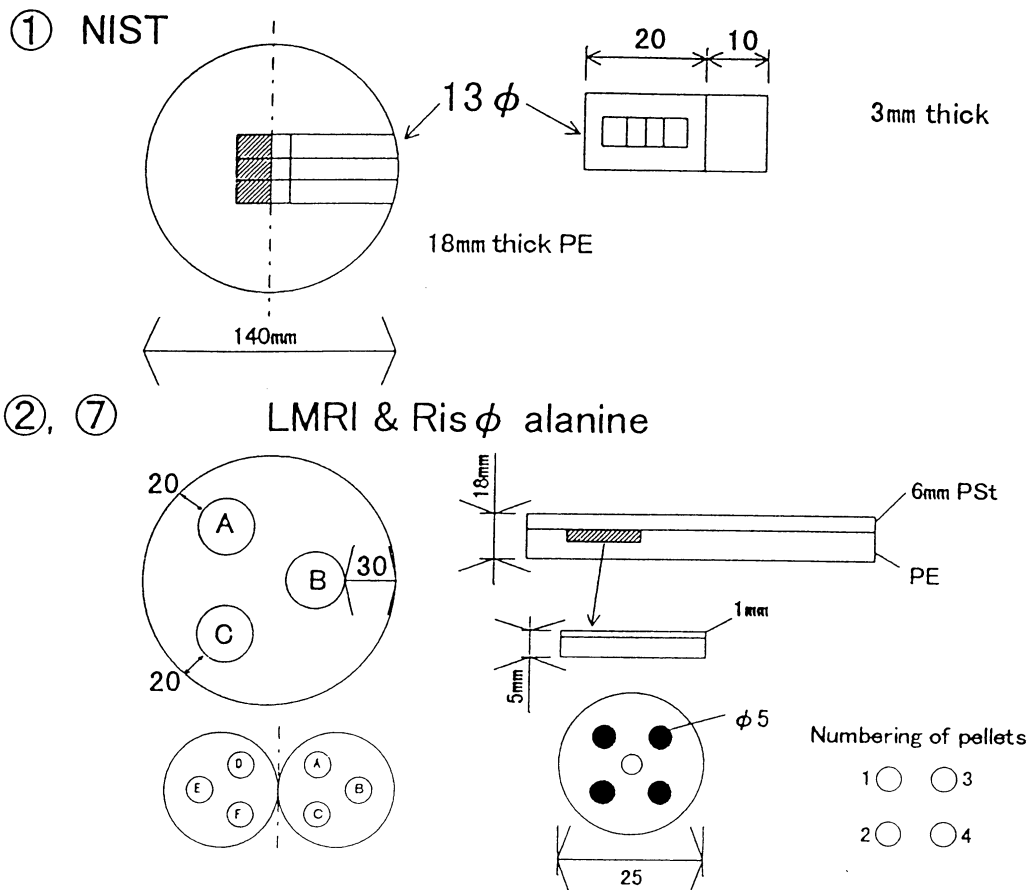


FIG. II.2. Schematic diagram showing the various irradiation phantoms in two trays (illustration by the courtesy of T. Satoh of JAERI).



③ IISC, ④ BARC, ⑤ IAEA/JAERI, ⑥ Nordion, ⑧ Ris  $\phi$  dichro same as ① except for hole diameter ( $\phi$  12)  
 Position of dosimeters was fixed by filling holes space by soft paper.

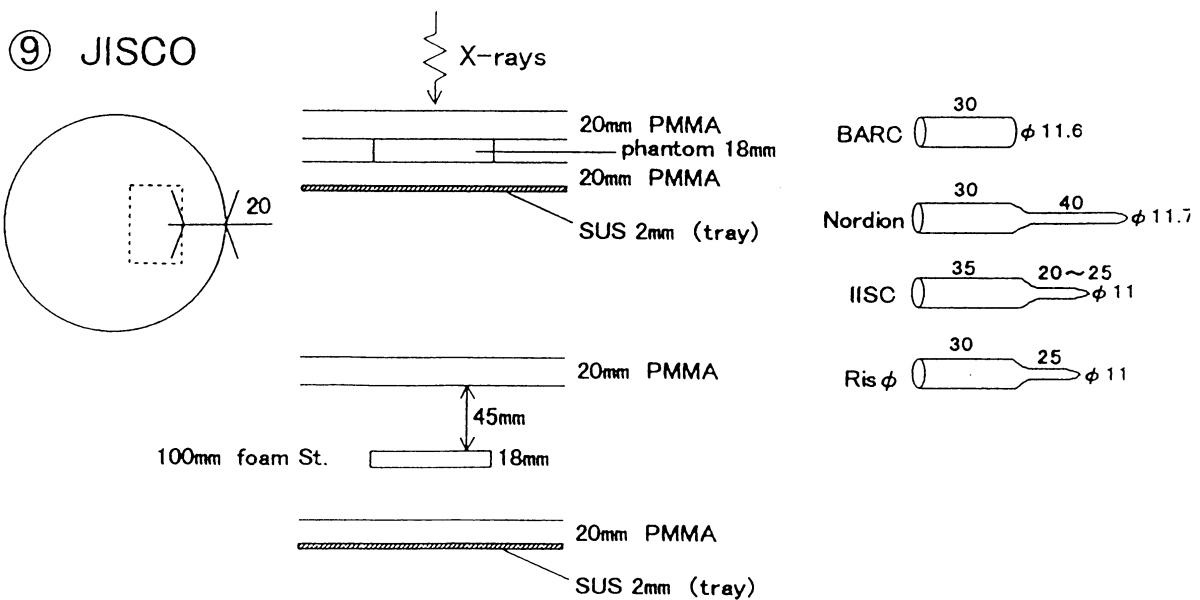


FIG. II.3. Details of dosimeter locations in the polystyrene irradiation phantoms (illustration by the courtesy of T. Satoh of JAERI).

These results are plotted in Fig. II.4, in which circles represent the ratio given in column 5 and crosses(x) indicate the three individual values. Bars show combined uncertainty values given in column 4.

TABLE II.3. RESULTS OF THE X RAY INTERCOMPARISON

ID Number	Dosimetry	Mean Dose (individual value)	Std uncertainty in mean (k = 1)	Lab/Mean
1	Alanine-ESR	4.15 (4.13, 4.16, 4.15)	1.6 %	0.99
2	Alanine-ESR	4.13 (4.09, 4.13, 4.16)	1.1	0.99
3	Alanine-ESR	4.26 (4.31, 4.22, 4.25)	1.6	1.02
4	Alanine-ESR	4.20 (4.19, 4.21, 4.19)	1.7	1.01
5	Alanine-ESR	4.09 (4.08, 4.08, 4.12)	1.8	0.98
6	Glutamine	4.03 (4.04, 4.01, 4.04)	2.0	0.97
7	Dichromate	4.31 (4.20, 4.32, 4.41)	2.3	1.03
8	ECB	4.22 (4.17, 4.30, 4.18)	2.5	1.01
9	Ceric-cerous	4.18 (3.93, 4.22, 4.15)	1.7	1.00
- MEAN OF	ALL LABS	4.17 ± 0.09 (2.1%)		

ID Number	Dosimetry	Mean Dose (individual value)	Std uncertainty in mean (k=1)	Lab/Mean
1	Alanine-ESR	8.24 (8.26, 8.27, 8.20)	1.6 %	1.00
2	Alanine-ESR	8.02 (7.97, 8.02, 8.08)	1.1	0.97
3	Alanine-ESR	8.43 (8.43, 8.37, 8.48)	1.5	1.02
4	Alanine-ESR	8.31 (8.34, 8.32, 8.27)	1.7	1.01
5	Alanine-ESR	7.95 (7.95, 7.90, 8.00)	1.8	0.96
6	Glutamine	8.05 (8.16, 8.00, 8.00)	2.0	0.97
7	Dichromate	8.62 (8.57, 8.63, 8.66)	1.2	1.04
8	ECB	8.35 (8.14, 8.45, 8.45)	2.5	1.01
9	Ceric-cerous	8.42 (8.40, 8.44, 8.41)	1.7	1.02
- MEAN OF	ALL LABS	8.27 ± 0.22 (2.7%)		

ID Number	Dosimetry	Mean Dose (individual value)	Std uncertainty in mean (k=1)	Lab/Mean
1	Alanine-ESR	12.49 (12.51, 12.53, 12.42)	1.6 %	1.01
2	Alanine-ESR	12.07 (12.04, 12.03, 12.15)	1.1	0.98
3	Alanine-ESR	12.54 (12.60, 12.35, 12.68)	1.9	1.01
4	Alanine-ESR	12.32 (12.39, 12.29, 12.29)	1.7	1.00
5	Alanine-ESR	12.10 (12.10, 12.00, 12.20)	1.8	0.98
6	Glutamine	12.09 (12.11, 12.11, 12.04)	2.0	0.98
7	Dichromate	12.69 (12.55, 12.66, 12.87)	1.6	1.03
8	ECB	12.57 (12.40, 12.60, 12.70)	2.5	1.02
9	Ceric-cerous	12.52 (12.56, 12.45, 12.55)	1.7	1.01
- MEAN OF	ALL LABS	12.38 ± 0.24 (1.9%)		

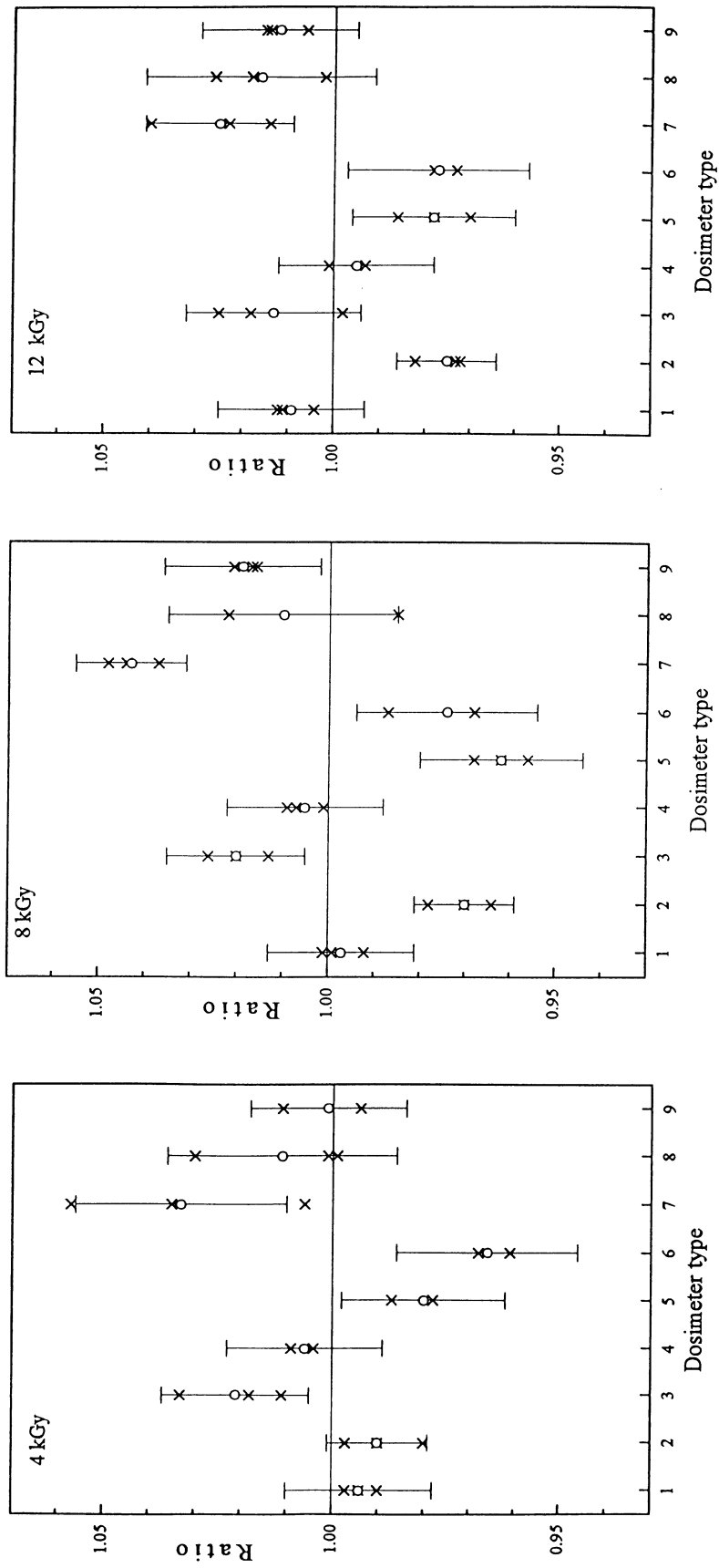


FIG . II.4. The circles(o) represent the ratio of laboratory value to the IAEA mean value (column 5, Table II.3), and crosses(x) represent the three individual values for a dosimeter set. Bars show combined uncertainty values as stated by each participant (column 4, Table II.3).

## Analyses

The standard deviation of all the 27 points is about 2.1%. The results are reasonably distributed; 17 points (63%) are within  $1\sigma$  from the mean, 9 points (33%) are within  $1\sigma$  and  $2\sigma$  from the mean, and one point (4%) is between  $2\sigma$  and  $3\sigma$  from the mean. This is confirmed by the very small difference between the arithmetic mean and the robust mean.

The uncertainty in the temperature value used for the correction of the temperature dependency of the dosimeter response could be about  $2^{\circ}\text{C}$ , which is equivalent to about 0.5% in the dose value. The uncertainty in the measured dose value due to the variation of dose rate both in lateral direction and along the beam is less than 2%. Almost all the dosimeters were in tightly screw-sealed plastic containers or flame-sealed glass ampoules until analyses, so that humidity effect may be negligible in this study.

Considering these influences as well as the uncertainties of the individual dosimetry systems, an agreement of 2.1% among the various dosimeters is within a reasonably achievable limit for high-dose intercomparison. This result is similar to the previous dose intercomparison study for  $^{60}\text{Co}$  gamma rays (Appendix I), although in that case one type of dosimeter, alanine dosimeters of IAEA, was irradiated at each participating laboratory.

All dosimetry systems were calibrated under  $^{60}\text{Co}$  gamma ray conditions. To check the influence of the energy spectrum on the performance of the dosimeters, the mass energy absorption coefficients weighted for entire energy spectra were estimated for 5-MV X rays, as shown in Table II.4 (Kojima). The coefficient values for PMMA and alanine dosimeters for  $^{60}\text{Co}$  gamma rays (1.25 MeV as average) and X rays with different energy spectra according to original electron energies are in good agreement. The results of intercomparison are consistent with these calculations.

TABLE II.4. EFFECTIVE MASS ENERGY-ABSORPTION COEFFICIENT ( $\text{cm}^2\text{g}^{-1}$ )

Material	1.25 MeV (Cobalt-60) gammas	5-MV X rays
Water	0.0297 (1.000)*	0.0282 (1.000)
Alanine	0.0288 (0.970)	0.0274 (0.972)
PMMA	0.0288 (0.970)	0.0274 (0.972)

\* The value in the parenthesis is the ratio of the coefficient to that of water.

## Conclusion

The above results suggest that all the dosimetry systems used in this study have a potential to be used for X rays produced by high-energy electron beams, when calibrated by using  $^{60}\text{Co}$  gamma rays.

## ACKNOWLEDGEMENTS

The IAEA would like to express its appreciation for all the assistance provided by West Japan Irradiation Service Co. Ltd to this dose intercomparison study for X rays.



## **Appendix III**

### **IRRADIATION DOSIMETER HOLDER**

#### **Introduction**

It is becoming quite apparent from various studies of influence quantities that it is essential either to calibrate the routine dosimetry system in the gamma irradiation plant (in-plant calibration) or at least verify the applicability of the calibration relationship under the conditions of use [4]. For this, it is recommended that the routine dosimeters and the reference (or transfer) dosimeters be irradiated in a standard holder that will ensure that the dosimeters receive essentially the same dose. This requirement also exists for the intercomparison of various dosimeters, like that described in Appendix II.

#### **Requirements**

The dosimeter holder should be placed in a region of low dose gradient, for example, in the middle of a homogeneous product. The wall should be thick enough to ensure that the dosimeters are surrounded with material which is similar to the dosimeter material in order to limit effects from interfaces. Recommended material is polystyrene or a similar radiation-resistant plastic with wall thickness of 5–8 mm to provide the electron equilibrium conditions. Increasing the wall thickness beyond this may create dose gradients within the phantom due to attenuation. Similarly, the mass of dosimeter material must not be so large that significant dose gradients are introduced — for two-sided irradiations the thickness of unit density material should not exceed 15 mm. For a one-sided irradiation larger uncertainties may be introduced due to attenuation. These may be reduced for two-sided irradiation. Generally, if the reference and routine dosimeters are arranged in a symmetrical "sandwich" along the direction of irradiation, no significant error will be introduced.

#### **Phantom design**

Examples of irradiation dosimeter holders for in-plant gamma irradiation are given in Fig. III.1. The upper phantom in the figure allows routine dosimeters (e.g. up to 5 PMMA) to be irradiated with a 6-mm thick alanine reference dosimeter on either side of them. The lower phantom can accommodate a larger rectangular insert intended to hold a dosimeter box containing routine dosimeters and two cylindrical reference dosimeters arranged in a line. This geometry is suitable for either liquid dosimeter ampoules or alanine holders.

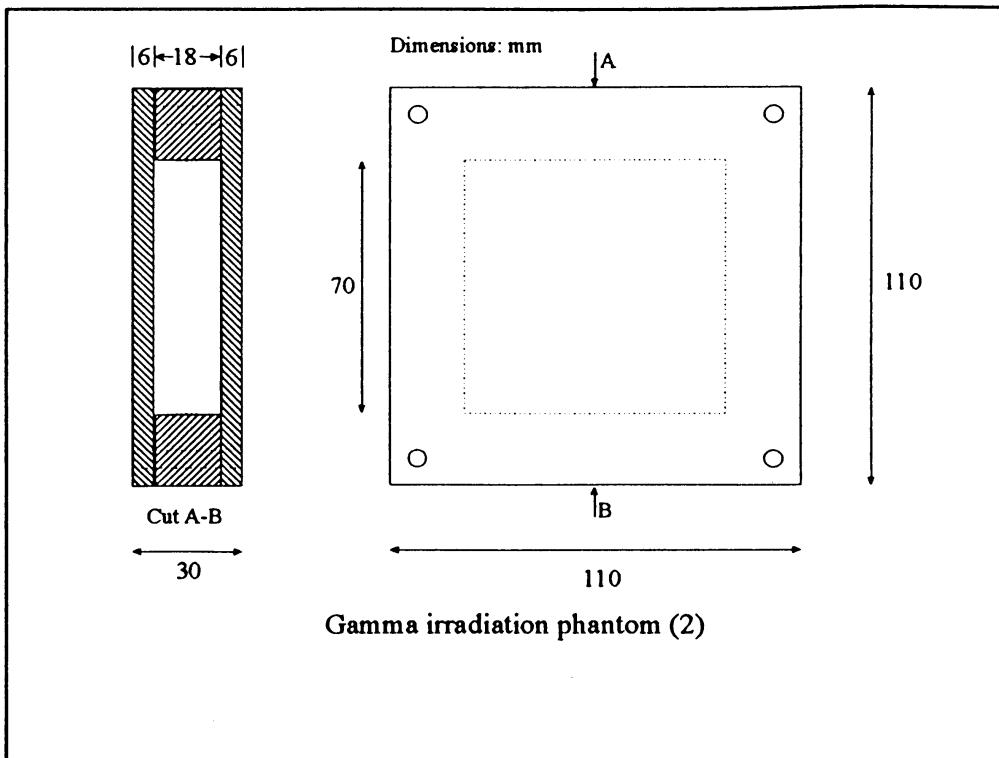
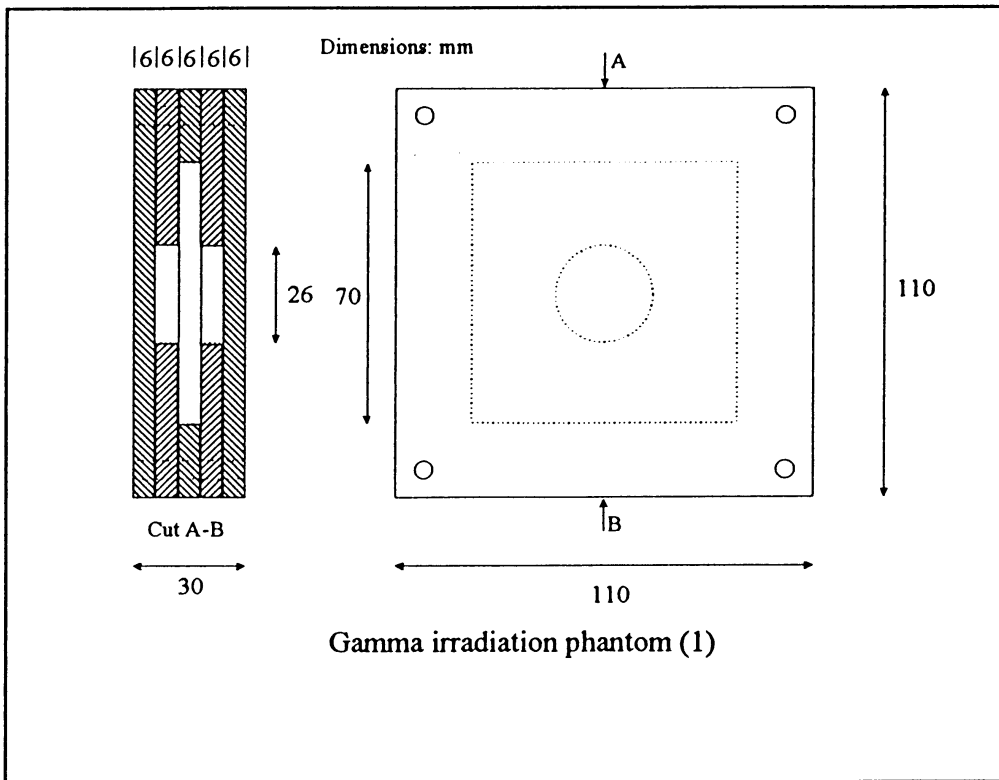


FIG. III.1. Two examples of dosimeter holders for in-plant irradiation.

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**Annex**

**FINAL REPORTS OF THE CRP PARTICIPANTS**



# COMBINED EFFECT OF TEMPERATURE, DOSE RATE, HUMIDITY AND ENERGY SPECTRUM ON THE RESPONSE OF COMMERCIAL ROUTINE DOSIMETERS USED IN INDUSTRIAL GAMMA PROCESSING

R. CHU  
MDS Nordion,  
Kanata, Ontario, Canada

## Abstract

Combined effects of different environmental factors can result in large uncertainties in the dose estimates obtained from the use of routine dosimeters in production  $^{60}\text{Co}$  irradiators. Studies were performed to determine the variation of dose rates and temperature in production irradiators and to reduce the uncertainty introduced by combined effects caused by the non-uniform rate of energy absorption. Methods for calculating dose rates were reviewed and calculated dose rates were compared with measured dose rates to provide a better understanding of the magnitude of the dose rate variations. The use of pairs of transfer standard dosimeters having approximately equal but opposite temperature coefficients were studied as a method to reduce the uncertainties in the dose estimate for the transfer standard dosimeters caused by uncertainties in the estimate of effective temperature.

## 1. INTRODUCTION

Environmental factors have a significant effect on the response of many commercial dosimeters used for industrial gamma processing. The ASTM guide for the selection and calibration of dosimetry systems [1] states that the combined effects of environmental factors may make it difficult to apply correction factors for the differences between the calibration conditions and the conditions encountered during use in production  $^{60}\text{Co}$  irradiators. To reduce uncertainties caused by combined effects, the ASTM guide recommends in-plant verification of the performance of a routine dosimetry system or in-plant calibration of the dosimetry system against a transfer standard dosimetry system.

MDS Nordion provides a dosimetry calibration service with irradiations performed under controlled temperature conditions in a gammacell 220 irradiator. It also provides a transfer standard dosimetry service using ceric-cerous dosimeters. MDS Nordion recommends in-plant calibration of routine dosimeters against transfer standard dosimeters and routinely performs this in-plant calibration during the installation qualification dosimetry of new production  $^{60}\text{Co}$  irradiators.

Although in-plant calibration provides a response function that minimizes uncertainties from environmental factors, this calibration method still has limitations. Factors that must be considered when placing pairs of routine and transfer dosimeters in the irradiator for in-plant calibrations include the effect of the high instantaneous dose rates during different portions of the irradiation cycle and the non-uniform temperature variation during and after the irradiation on the transfer dosimeter response. Some locations may have large dose gradients requiring precise location of the dosimeter pairs. Detailed knowledge of the dose rate and temperature variation as the dosimeters move through the irradiator is required to ensure that the dose estimate obtained from the calibration response function is valid for the actual irradiation conditions.

The uncertainty contribution from the combined effects of dose rate and temperature on the response of routine dosimeters caused by this non-uniform rate of energy absorption is unknown and is ignored in the uncertainty estimate or an arbitrary estimated value is assumed.

Better understanding of the rate of the increase in dose and temperature and the effect on routine dosimeters will help in the optimization of the radiation process.

The studies performed under this research agreement address these issues related to combined effects in production  $^{60}\text{Co}$  irradiators. Although relative humidity and changes in energy spectrum also introduce errors in the dose estimate, the studies focused on the combined effects of dose rate and temperature. For routine dosimeters that can be affected by relative humidity, it is assumed that they have been conditioned under controlled conditions to constant levels of water content and packaged in hermetically sealed containers. Suppliers of routine dosimeters are encouraged to supply these prepackaged dosimeters for use in gamma irradiators if relative humidity changes are found to have any effect.

For dosimeters containing materials with higher atomic number, the energy absorption may be affected by the lower energy photons in different portions of the irradiation cycle. The work on Monte Carlo calculations using the MCNP code that was started during this study will provide a method for obtaining the energy spectrum in production  $^{60}\text{Co}$  irradiators that may help assess this effect on the dosimeter response.

## 2. COMBINED EFFECTS IN PRODUCTION $^{60}\text{Co}$ IRRADIATORS

A major factor contributing to the uncertainty of routine dose measurements in production  $^{60}\text{Co}$  irradiators is the wide variation in the rate at which energy is absorbed. The rate at which energy is absorbed in a dosimeter varies as the product moves through the irradiator and also depends on the location of the dosimeter in the product. For some tote-type irradiators, product is irradiated on a lower level and then raised to an upper level for completion of the irradiation. In these irradiators, some regions near the top of the product may receive more than 50% of the total dose at one or two irradiation positions during the first quarter of the irradiation cycle while regions near the bottom of the product on the other side of the tote may not receive this same high dose-rate contribution until the final quarter of the irradiation cycle.

At present, most of the information available on dose rate variations in industrial irradiators is obtained from calculations based on the point kernel method. Some dosimetry data had been obtained from static dose rate measurements or from the irradiations of dosimeters for portions of the irradiation cycle. The Real Time Dosimeter [2] developed by the UK National Physical Laboratory (NPL) provides a new method for routine dose measurements. Although not a direct part of this CRP, the NPL Real Time Dosimeter was used to measure the dose rates at several production  $^{60}\text{Co}$  irradiators and the measured dose rates were compared with dose rate calculations for the individual irradiation positions.

During the irradiation, the temperature of the dosimeter increases as a result of the energy absorption in the dosimeter and in the surrounding products and containers. This temperature increase depends on the specific heat of the materials and the total integrated dose. The rate of temperature increase is also influenced by contributions from radiant heat from the  $^{60}\text{Co}$  sources and radiation and conduction from metal structures. At some portions of the product, cooling caused by air flowing by the source and product may reduce the heat contribution and result in only a small temperature increase. For dosimeters in products with good insulating properties, the dosimeter temperature may remain near the maximum value for hours after the completion of the irradiation. There may be wide variations in the



temperature profile between summer and winter, especially if the summers are hot and the air used for ventilation of the irradiation room is not cooled.

The temperature profile is difficult to calculate and most of our knowledge of the rates of temperature increase has been obtained using mechanical recording bimetallic thermometers. The maximum temperatures during irradiation are fairly well known since they can be routinely measured using temperature-indicating labels that change colour when different temperature increments are reached.

Harwell Red 4034 and Far West Technology FWT-60-00 dosimeters are the most common dosimeters used for absorbed dose mapping or routine monitoring in production  $^{60}\text{Co}$  irradiators. Since the response of both of these dosimetry systems may be affected by combined effects of dose rate and temperature a major focus of our research was to reduce the uncertainty in dose measurements using these systems caused by dose rate or temperature effects. Red 4034 dosimeters are always supplied in hermetically sealed sachets, so effects of relative humidity are a concern only if the sachets are damaged or if the sachets are opened to obtain pre-irradiation readings. FWT-60-00 dosimeters are supplied in hermetically sealed packages or as sets of several dosimeters in hermetically sealed sachets. For measurements in gamma irradiators, users are encouraged to use conditioned FWT-60-00 dosimeters packaged in hermetically sealed sachets.

During the studies, efforts were also made to find alternative superior dosimetry systems suitable for routine dose measurements. Studies were performed with developers of new dosimetry systems to determine if any of the new systems would provide advantages over the currently used systems with less influence from combined effects.

### 3. COMPARISON BETWEEN CERIC-CEROUS AND RED 4034 DOSIMETER READINGS

During the installation qualification of all new industrial irradiators built by MDS Nordion, installation qualification dose mapping is performed using ceric-cerous dosimeters as the reference standard dosimeter. By including the customer's routine dosimeter with the ceric-cerous dosimeters, in-plant calibration against the ceric-cerous dosimetry system is also performed at this time. Some variations are usually seen between the ceric-cerous doses and the doses obtained from the routine dosimeters calibrated by irradiations at a calibration facility. However, because of the limited knowledge provided to MDS Nordion on the dosimeter and instrument calibration, it is difficult to assess what difference may be caused by influence quantities.

To determine possible effects from influence quantities on Harwell Red 4034 dosimeters, MDS Nordion provided Red 4034 dosimeters with ceric-cerous dosimeters in the calibration packets used for the dose mapping studies at the commissioning of a number of irradiators. The Red 4034 dosimeters were calibrated by irradiation in the same irradiation facility as the ceric-cerous dosimeters so systematic differences in the calibration were eliminated. The Red 4034 dosimeters were returned to MDS Nordion for reading using the same spectrophotometer as used in the calibration so, again, systematic errors due to instrument calibrations were eliminated.

Table I compares the ceric-cerous dosimeter readings with the readings of Harwell Red 4034 dosimeters supplied and read by MDS Nordion for ten production irradiators. Both of

the dosimetry systems were calibrated by irradiations in the same gammacell 220 at a temperature of 25°C so any systematic errors between calibration facilities are eliminated.

The results given in Table I show fairly good agreement between the ceric-cerous and the Harwell Red 4034 readings. In 7 out of the 10 cases examined, the difference was 3% or less. The largest difference was 8%. These results indicate that the response functions obtained from calibrations by irradiation in a calibration facility often give dose values in good agreement with the dose values that would be obtained from an in-plant calibration performed at the production irradiator. However, these results and results from other studies do not show trends that indicate when differences may be expected to occur. Studies performed under this CRP may provide insight for determining better methods for predicting when deviations will occur.

TABLE I. COMPARISON BETWEEN CERIC-CEROUS AND HARWELL RED 4034 DOSES MEASURED IN PRODUCTION IRRADIATORS BASED ON CALIBRATIONS IN GAMMACELL 220

Irradiator	Irradiator Type	Activity (MCi)	Red 4034/Ceric-Cerous
1	Carrier	0.7	1.08
2	Carrier	0.4	1.02
3	Tote	0.5	1.04
4	Carrier	1.0	1.05
5	Carrier	1.6	1.02
6	Tote	1.1	0.98
7	Carrier	1.4	1.02
8	Pallet	0.7	1.02
9	Carrier	1.2	1.00
10	Tote	3.1	0.97

#### 4. TEMPERATURE-INDEPENDENT MEASUREMENTS

At the commencement of the research project, MDS Nordion was working with Bruker Instruments, Inc. on the potential for use of the alanine dosimetry system as a superior routine dosimetry system. The use of alanine dosimeters as a routine dosimeter could be justified if the higher cost of the EPR spectrometer was offset by reduced measurement uncertainties resulting in increased throughput and reduction in the <sup>60</sup>Co requirements. The use of alanine dosimeters as a transfer standard dosimeter could also complement the transfer standard dosimetry service now being offered using ceric-cerous dosimeters. The wide dose range of the alanine dosimetry system would allow expansion of the transfer dosimetry service beyond the range presently covered by ceric-cerous dosimeters.

The irradiation temperature affects the response of most reference dosimeters, including ceric-cerous, alanine and dichromate. The temperature coefficient for these systems is approximately 0.2%/°C (a small effect), and corrections are usually made based on a

measurement of the maximum irradiation temperature. However, because of the uncertainties in the rate of temperature increase, the uncertainty of the temperature correction can be significant.

Since the MDS Nordion ceric-cerous dosimetry system has a negative temperature coefficient of approximately  $-0.2\%/^{\circ}\text{C}$  and published data on alanine dosimetry systems showed a positive temperature coefficient of approximately  $+0.2\%/^{\circ}\text{C}$ , the use of pairs of ceric-cerous and alanine dosimeters appeared to be a method for obtaining temperature independent measurements without the need for a precise knowledge of the temperature profile during the irradiation.

During the research project, MDS Nordion decided to stop further work with Bruker Instruments Inc. on the use of alanine dosimeters for routine dosimetry. However, the determination of the advantages of a temperature-independent transfer dosimeter-pair still appeared to be a worthwhile goal. Hitachi Aminogray alanine dosimeters prepared for the International Dose Assurance Service (IDAS) [3] were provided by the IAEA for these studies.

At the three Allegiance irradiators in El Paso, Texas, routine dosimetry had been performed using FWT-60-00 dosimeters. However, wide variations in the dosimeter readings resulted in increased cycle times, loss of product throughput and high measured ratios of maximum to minimum dose. Interest in improved dosimetry resulted in a decision to use ceric-cerous dosimeters for routine dosimetry

For the ceric-cerous dosimetry system, the only significant influence quantity is the irradiation temperature. However, because of the uncertainty of the effective temperature to be used when correcting for the irradiation temperature, conservative values for the temperature correction factors are being used. Improvement in the knowledge of the effective temperature has significant economic impact, since a reduction in the  $^{60}\text{Co}$  activity may be possible. Although the activity reduction as a percent of the total activity is small, the actual value is significant because of the large quantity of  $^{60}\text{Co}$  at this site.

The use of pairs of ceric-cerous and alanine transfer standard dosimeters to eliminate the requirement to apply a correction factor for the irradiation temperature was studied at the three Allegiance irradiators. As part of this study, a number of measurements were performed using both temperature indicator labels and recording thermometers to determine the temperature profile for these irradiators. Several detailed dose maps, including a partial dose map at one irradiator to obtain information on the dose rates at the different irradiation positions, had previously been performed at these irradiators.

Because the difference in the response of ceric-cerous and alanine dosimeters in production irradiators expected from the difference in the temperature coefficients was small, studies were also performed to confirm that there were no systematic errors causing measurement differences. These studies consisted in the irradiations of pairs of alanine and ceric-cerous dosimeters together in a temperature-controlled gammacell 220 (GC 220) at MDS Nordion and an Underwater Calibrator (UWC) without temperature control at Allegiance. The results given in Table II showed good agreement at  $25^{\circ}\text{C}$  and gave differences at higher temperature close to those expected based on the known temperature coefficients.

TABLE II. COMPARISON BETWEEN CERIC-CEROUS AND ALANINE READINGS FOR IRRADIATIONS IN CALIBRATION FACILITIES AND EVALUATION AT AN IRRADIATION TEMPERATURE OF 25°C

Irradiation Facility	Temperature °C	Ceric-Cerous Dose 25, 35, 45 kGy	Alanine Dose 25, 35, 45 kGy	Alanine/Ceric-Cerous
GC 220	25	25.24, 35.34, 45.42	25.36, 36.10, 45.98	1.005, 1.002, 1.012
GC 220	44	23.78, 33.65, 43.28	26.88, 38.77, 49.45	1.130, 1.152, 1.143
UWC	25–28	24.58, 33.36, 44.30	25.13, 33.83, 45.98	1.022, 1.014, 1.038

The readings from the pairs of dosimeters irradiated in the three Allegiance production irradiators and evaluated at an irradiation temperature of 25°C are shown in Table III. For these irradiations, four ceric-cerous dosimeters and a cylinder containing three alanine dosimeters were placed in a plastic holder designed to hold five pharmaceutical ampoules. This holder ensured that all dosimeters were precisely located and received the same dose. A mechanical recording thermometer with a bimetallic sensor and a wind-up spring-operated drive to record the readings on a circular chart was placed with the dosimeters to measure the irradiation temperature. The effective temperature was estimated from the average of the maximum temperature measured by the recording temperature and the ambient temperature prior to the start of the irradiation. The effective temperatures calculated from the average of the ceric-cerous and alanine dose values are compared to the temperatures calculated from the readings of the recording thermometer are shown in Table IV.

TABLE III. READINGS FROM PAIRS OF CERIC-CEROUS AND ALANINE DOSIMETERS IRRADIATED IN PRODUCTION IRRADIATORS BASED ON EVALUATIONS AT 25°C IRRADIATION TEMPERATURE

Irradiation Facility	Position (Min or Max)	Ceric-Cerous Dose (kGy)	Alanine Dose (kGy)	Average Dose (kGy)
Unit 1	Min	37.98	42.44	40.21
	Max	39.80	45.07	42.44
Unit 2	Min	24.33	25.58	24.95
	Max	25.95	27.67	26.81
Unit 3	Min	25.83	26.91	26.37
	Max	27.90	29.71	28.81

TABLE IV. COMPARISON BETWEEN ESTIMATIONS OF EFFECTIVE TEMPERATURES

Irradiation Facility	Minimum or Maximum Dose Position	Temperature Estimate from Recording Thermometer (°C)	Temperature Estimate from Dosimeter Pairs (°C)	$\frac{\text{Temp Rec Therm (°C)}}{\text{Temp Dos Pairs (°C)}}$
Unit 1	Min	46.5	48.5	0.96
	Max	46.5	51.3	0.91
Unit 2	Min	37.5	35.7	1.05
	Max	37.5	38.6	0.97
Unit 3	Min	35.0	33.7	1.04
	Max	35.0	38.3	0.91

These two temperature estimates are in good agreement. Some uncertainty is introduced by the precision of the dosimeter measurements, but this uncertainty can be reduced by an increase in the number of measurements. The method appears to be very promising for reducing the uncertainties in the in-plant calibration procedure caused by the estimation of the effective irradiation temperature. An improvement of 10°C in the estimate of the effective temperature will reduce the uncertainty by 2%.

## 5. IRRADIATOR DOSE RATE MEASUREMENTS AND CALCULATIONS

Point kernel calculations have been performed by MDS Nordion for many years and the calculated results are routinely compared against dosimetry measurements. However, to understand the dose rate variation in the irradiator the actual dose rates at the different irradiation positions must be well known. Because there are often correction factors applied based on comparison with dosimetry, the accuracy of the modelling methods for obtaining estimates of dose rates has only been validated for limited conditions. Measurements of dose rates in production irradiators are possible with integrating dosimeters, but the performance of these measurements without undue interruption of commercial operations is difficult without real time measurements.

Calculated dose rates for production  $^{60}\text{Co}$  irradiators have been obtained from computer models using the point kernel method. During the research agreement, a thorough review was performed on the computer model now used by MDS Nordion. Validation studies were performed to confirm the consistency of present calculations with calculations performed using previous computer models and with total integrated doses obtained from dose mapping measurements. Work was also started on the use of the MCNP Monte Carlo code to obtain an improved model of the absorption and scattering of  $^{60}\text{Co}$  gamma rays passing through the different materials in the product, source and irradiator mechanism.

Measurement data on dose rates at the different irradiation positions had previously been obtained from static irradiation studies or by the irradiation of dosimeters for different fractions of the irradiation cycle. Direct measurements of dose rate obtained from studies with the NPL Real Time Dosimeter provided data for the individual dwell positions for comparison with dose rates obtained from point kernel calculations. The NPL Real Time Dosimeter also provided temperature measurements for the dosimeter position for different fractions of the irradiation cycle.

The NPL Real Time Dosimeter now provides a direct means for dose rate measurements for comparison with calculations. Figure 1 shows the dose rates at the side of a tote box in a JS7500 Tote Box Irradiator calculated using the point kernel method compared with the dose rates measured using the NPL Real Time Dosimeter. Differences observed at different portions of the irradiation cycle are being reviewed to determine if changes can be made to the computer calculations to improve the agreement. Monte Carlo calculations will also be used to assess possible causes for the differences.

## 6. PERFORMANCE QUALIFICATION TESTING OF DOSIMETRY SYSTEMS

At the International Symposium on Techniques for High Dose Dosimetry in Industry, Agriculture and Medicine, Vienna, 2–5 November 1998 the author presented an invited paper on the effects of influence quantities on radiation processing dosimetry [4]. During the

discussions following this presentation it became clear that there was an urgent need for standard methods for testing new dosimetry system to systematically examine effects of different environmental factors. Studies of the effects of individual influence quantities will not necessarily determine the combined effect when the dosimeter is used in a production irradiator since there may be synergistic relationships between some of the influence quantities. However, it was felt that systematic performance testing would alert the developers of new dosimetry systems of these effect at an early stage of the dosimetry system development so that efforts could be directed towards finding changes that could reduce them.

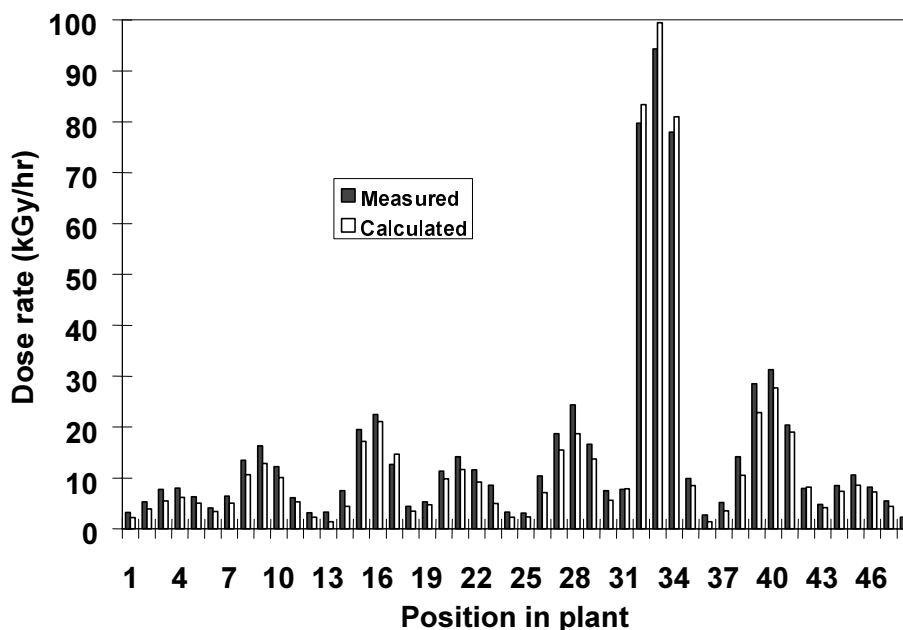


FIG. 1. Comparison between NPL measured dose rates and dose rates calculated by point kernel method for JS 7500 tote type irradiator.

As a result of these discussions, the author volunteered to chair an ASTM Task Group to write a standard for performance testing of dosimetry systems. Although this work is not directly a part of this CRP, the standard will make the radiation processing industry more aware of the uncertainties introduced by influence quantities.

## 7. DISCUSSION

The result of this research are very relevant to the first objective of the CRP, which was to understand and evaluate the influence of various external parameters on the performance of several routine dosimeters in use at the present. The study focused on two parameters, dose rate and temperature, whose combined effects are a major source of dosimetry uncertainty. Data obtained from this study will result in improved calibration techniques resulting in increased accuracy and reliability of the routine dosimetry systems.

Work performed under this research agreement was not expected to be relevant to the development of reference dosimetry techniques for electron beams of energy less than 4 MeV.

However, the results are applicable for X ray sources where effects of influence parameters are expected to be similar to those for gamma sources. The broad energy spectrum of the photons generated by X ray sources is an additional potential source of error that can be assessed using the Monte Carlo programme being developed for calculating dose rates and energy distributions in production  $^{60}\text{Co}$  irradiators.

### ACKNOWLEDGEMENTS

Much of the work performed for this research project was performed in co-operation with operators of MDS Nordion irradiators or with suppliers of dosimeters. We especially want to thank Adriana Uranga of Allegiance Healthcare Corporation for her assistance in providing measurement data from the three production  $^{60}\text{Co}$  irradiators operated by Allegiance in El Paso, Texas. Adriana performed detailed temperature measurements at the three irradiators using recording thermometers and temperature indicators to increase our knowledge of the temperature profile in industrial irradiators. She also performed the irradiation of pairs of ceric-cerous and alanine dosimeters for the studies to obtain temperature-independent dosimetry.

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# CHARACTERIZATION AND EVALUATION OF PVG DOSIFILM FOR LOW ENERGY ELECTRON BEAM

W. CHEN, H. JIA

Radiation Chemistry Group,  
Radiochemistry and Radiation Chemistry Division,  
Department of Chemistry, Beijing Normal University,  
Beijing, China

## Abstract

Experiments have shown that the PVG dosifilm can be used as a routine dosimeter for gamma rays and electron beam irradiation. The specific net absorbance of the irradiated PVG dosifilm is affected by the irradiation temperature and relative humidity. The average value of the irradiation temperature coefficient in the temperature range 18 to 55°C was found to be 0.008 of the response/°C. The relative humidity effect on the irradiated PVG dosifilm can be neglected below 30% r.h., but the correction is necessary at higher relative humidity. The average value of the coefficient is 0.006 of the response/% of r.h. in the range of 30% to 75% r.h. The irradiated PVG dosifilm must be stored for at least 24 hours after irradiation before reading due to the post-irradiation effect. The total uncertainty in the dose measured by the PVG dosifilm is less than 10% for EB irradiation.

## 1. INTRODUCTION

The absorbed dose is an important parameter for the quality control of radiation processing. With the growing interest in the use of electron beam processing, more attention was drawn to the dose measurement for EB and more new requirements for quality control by dosimetry were recognised. Some investigation of dosimeters for low energy electron beam (<10 MeV) was carried out. Plastic dosifilm is suitable for radiation processing as a routine dosimeter with its reasonable cost for the convenience of dose mapping and for the possibility of file keeping. Its density is close to water and tissue, this makes it suitable for dose measurements in a widespread materials. Dosifilm is particularly suited to the EB dose measurement due to its small size. Present report is an investigation of the radiochromic PVG dosifilm for absorbed dose measurement of low energy EB irradiation. A comparison of dose responses for gamma rays and EB irradiation was carried out, meanwhile various effects (including the irradiation temperature, relative humidity, post-irradiation effect, dispersion and reproducibility) were tested and discussed.

## 2. EXPERIMENTAL

### 2.1. PVG dosifilm

Suppliers:

Leuco-malachite green (LMG, SIGMA Co., USA)

Polyvinyl butyral (Tianjin Organic Chemistry Industrial Experimental Factory, China)

Alkyl halide (RX, AR, Tianjin, China).

Composition:

Content of LMG in PVB — 80  $\mu\text{mol/g}$

The ratio of mass of RX and LMG — 0.5.

The above materials were dissolved in mixed solvent (alcohol, water, ethyl acetate, etc.). Films were cast by pouring the above solution on glass surface and dried. The thickness of dried film was  $40 \pm 5 \mu\text{m}$ .

The absorbance of dosifilm was measured by DMS-300 spectrophotometer (Vrian Ltd, USA) at least one day after irradiation. Dosifilm thickness was measured by Elecont micrometer Mitutoyo, JAPAN with a reproducibility of  $\pm 0.2 \mu\text{m}$ .

## 2.2. Irradiation

Cobalt-60 gamma ray facility with activity  $5.5 \times 10^{14}$  Bq at BNU. The absorbed dose rate was measured by Fricke and dichromate dosimeters.

BF-5 LINAC, Institute of Low Energy Physics, BNU, energy 3–5 MeV (variation  $\pm 5\%$ ), current intensity  $100 \mu\text{A}$ , pulse length  $2 \mu\text{s}$ , pulse frequency 500 pps, scan width 60 cm. The absorbed dose was calibrated by a calorimeter and dichromate dosimeters for EB irradiation. In order to approximate electron equilibrium conditions the PVG dosifilms were irradiated in a polystyrene phantom for gamma rays and in a polystyrene water jacket for EB to maintain constant temperature.

A water jacket with recycle water from a thermostat was used for keeping a given constant temperature during irradiation. The temperature was read out from both thermometers, which were put in the inlet and outlet of water jacket. The temperature difference between both thermometers for each given temperature was less than  $0.5^\circ\text{C}$  during irradiation. In front of dosifilm, the thickness of wall material (plastic) of water jacket plus the thickness of water layer facing the electron beam was 10 mm.

Several plastic vials with various relative humidities were prepared for PVG dosifilms irradiation. The relative humidity was 0%, 33.6%, 54.9% or 75.7% in each vial. Three PVG dosifilms were placed in each vial at least one day before irradiation.

## 3. RESULTS AND DISCUSSION

### 3.1. General characterization of PVG dosifilm

There are two absorption peaks (425 nm and 627 nm) in the spectrum of the irradiated PVG dosifilm. The absorbance values of the irradiated PVG dosifilm at these wavelengths increased with increasing absorbed dose as shown in Fig. 1.

From the gamma ray curve (■), it can be seen that in the range of 5 to 100 kGy, the specific net absorbance ( $\Delta A/\text{Thickness (mm}^{-1}\text{)}$ ) increased with the increase of dose with the sensitivity of 1.065. However, the absorbance was over 3 (transmission 0.001%) above 80 kGy, and it was difficult to increase beyond this. The electron curve (●) shows that the sensitivity of PVG dosifilm irradiated by EB was 0.783 up to 150 kGy. The relationships of the specific net absorbance vs. absorbed dose for PVG dosifilm irradiated at  $20^\circ\text{C}$  by gamma rays and by EB are as follows:

$$\Delta A/\text{Thickness (mm}^{-1}) = 2.40 + 1.065D - 0.002D^2 \dots \text{ for cobalt-60 gamma rays (1)}$$

$$\Delta A/\text{Thickness (mm}^{-1}) = 0.56 + 0.783D - 0.002D^2 \dots \text{ for 3-5 MeV electrons (2)}$$

where, D is absorbed dose (kGy).

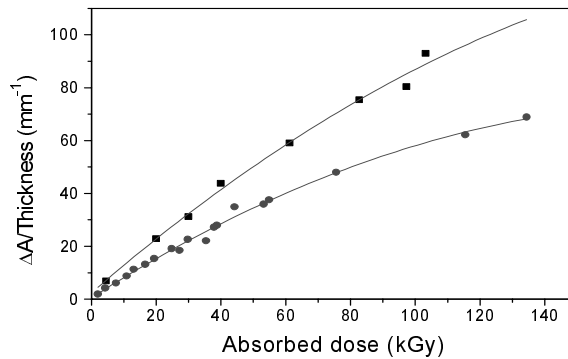


FIG. 1. Specific net absorbance ( $\Delta A/\text{Thickness (mm}^{-1})$ ) vs. absorbed dose for PVG dosifilm irradiated by cobalt-60 gamma rays (■) and by EB(●).

### 3.2. Influence of some factors on measurements of irradiated and unirradiated PVG dosifilm

#### 3.2.1. Colour development of PVG dosifilm irradiated by gamma rays and EB

There was post-irradiation effect for PVG dosifilm irradiated by both gamma rays and EB, i.e. the colour of the irradiated PVG dosifilm was built up over a period of time. Figure 2 shows the colour built-up time at 627 nm for PVG dosifilm irradiated by gamma rays at room temperature. The full colour built-up time of PVG irradiated by gamma rays at 15 kGy was about 20 h. The full colour built-up time of PVG dosifilm irradiated by EB at 21.6 kGy and 65 kGy increased from 22 to 24 h (see Fig. 3).

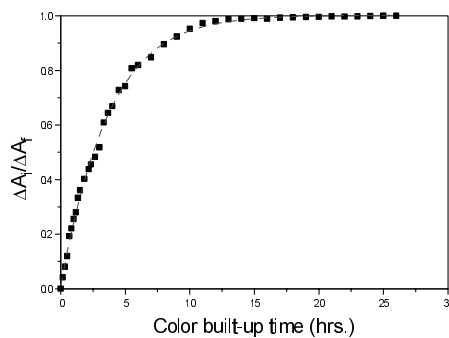


FIG. 2. The colour built-up time for PVG irradiated by  $\gamma$  rays, absorbed dose 15 kGy.

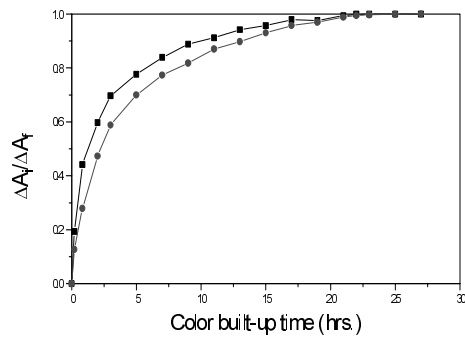


FIG. 3. The colour built- up time for PVG dosifilm irradiated by EB at different absorbed doses. ■ -21.6 kGy ● -65 kGy.

where,

$\Delta A_i$  is the net absorbance of PVG dosifilm immediately after irradiation.

$\Delta A_f$  is the net absorbance of PVG dosifilm later after irradiation.

Figure 4 shows the colour built-up time of PVG irradiated by EB at different storage temperatures. The colour built-up time can be accelerated by increasing the storage temperature, i.e. the colour development rate depends on the storage temperature. Figure 5 shows the relationship of full colour built-up time vs. storage temperature. It indicates that the full colour built-up time is shortened at a rate of 0.4 to 0.5 h/°C at the same absorbed dose. Generally, the irradiated PVG dosifilm must be stored at least 24 h after irradiation at room temperature in order to ensure the reproducibility of results.

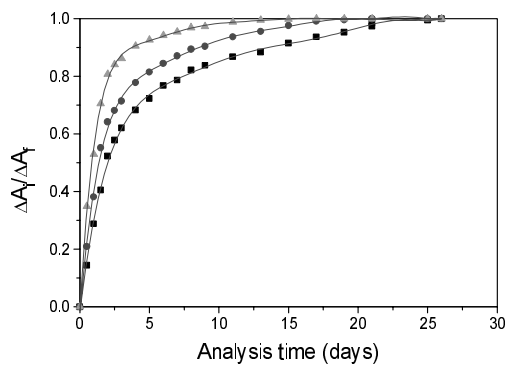


FIG. 4. Colour built-up for PVG dosifilm, 65 kGy, at different storage temperatures; ■ -18.5 °C, ● -35 °C, ▲ -45 °C.

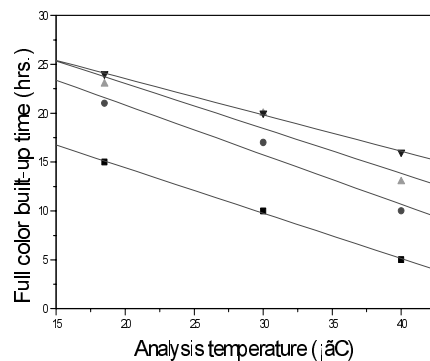


FIG. 5. Relationship of colour development vs. storage temperature for PVG dosifilm at various doses. ■ –8.6 kGy, ● –21.6 kGy, ▲ –28 kGy, ▼ –65 kGy.

### 3.2.2. Comparison between the decay of ESR spectrum and the colour development of PVG dosifilm

The PVG dosifilm, pure PVB, pure LMG, pure RX powder and mixture RX+LMG were irradiated by EB, and the ESR signal measured immediately by JEOL-1X ESR spectrometer to find the source of the  $RX^\bullet$  radical signal that appears in the ESR spectrum.

As shown in Table I, the pure PVB and pure LMG have no ESR signal, while the pure RX and mixture RX+LMG have a quartet peak (each peak line width 15 G) with the ESR signal intensity ratio 1:3:3:1. There was a singlet peak with line width about 60 G on ESR spectrum of PVG dosifilm. The singlet peak of ESR signal of PVG dosifilm should be an overlap spectrum, which consisted of  $RX^\bullet$  with another long-life radical signal. The colour built-up of PVG dosifilm should be related to the transient species  $RX^\bullet$  from RX, then  $RX^\bullet$  oxidized LMG to  $MG^+$ .

TABLE I. ESR SPECTRA OF IRRADIATED PVG DOSIFILM AND OTHER MATERIALS

Irradiated samples	ESR signal
PVG dosifilm	
Pure PVB	no ESR signal
Pure LMG	no ESR signal
Pure RX	
RX + LMG	

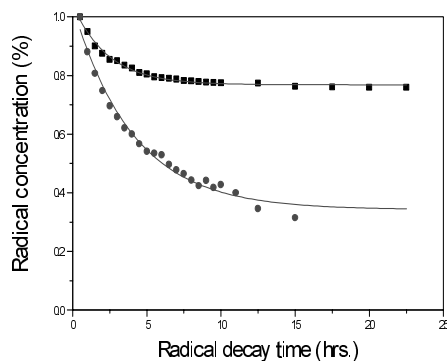


FIG. 6. Decay of radical of irradiated PVG dosifilm and of RX + LMG ■— PVG, ●— RX + LMG.

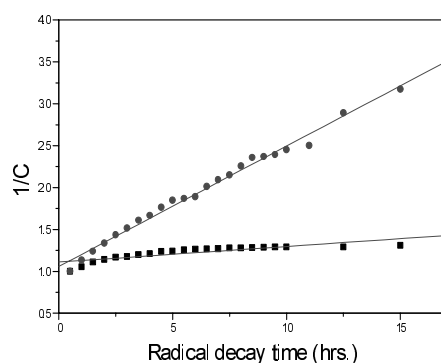


FIG. 7. Reciprocal of concentration of radical  $RX^\bullet$  ( $1/C$ ) of PVG dosifilm and of RX + LMG system vs. radical decay time. ■— PVG, ●— RX + LMG.

The radical decay curves for the irradiated PVG dosifilm and the RX + LMG system are shown in Fig. 6. The decay period of the radical is about 15 to 20 h. This decay period coincides with the full colour built-up time (15 to 20 h) of the irradiated PVG dosifilm. Figure 6 indicates that the final concentration (about 8%) of the radical of the irradiated PVG dosifilm (curve ■) is higher than that of RX + LMG (about 4% on curve ●). This may be related to the fact that a portion of  $RX^\bullet$  radical did not oxidize to LMG to  $MG^+$ , but transferred to a long life radical ( $RX^\bullet$ —polymer), which should be a radical linked with polymer. The reaction of  $RX^\bullet$  with LMG or with polymer as follows:



Figure 7 shows the reciprocal of the radical relative concentration ( $1/C$ ) vs. decay time (h) of the RX + LMG system and the irradiated PVG dosifilm. Both curves are linear i.e. the decay of  $RX^\bullet$  either from RX + LMG or from PVG dosifilm follows a secondary kinetic reaction.

The full colour development and radical decay period showed that the readout of PVG dosifilm was suitably taken at least one day after irradiation.

### 3.2.3. Effect of irradiation temperature on PVG dosifilm

Figure 8 shows the specific net absorbance of PVG dosifilm vs. absorbed dose irradiated by EB at various temperatures. Absorbed dose was calibrated by dichromate dosimeter and calorimeter. According to Eq. (2), the absorbed dose (D) for the PVG dosifilm irradiated by EB can be calculated from the specific net absorbance ( $\Delta A/\text{Thickness} (\text{mm}^{-1})$ ).

Curves with symbol  $\bullet$  ( $35^\circ\text{C}$ ),  $\blacktriangle$  ( $45^\circ\text{C}$ ) and  $\blacktriangledown$  ( $55^\circ\text{C}$ ) in Fig. 8 show that at the same dose the specific net absorbance of the irradiated PVG dosifilm increased with increasing irradiation temperature. Specific net absorbance of PVG dosifilm vs. irradiation temperature for various doses are shown in Fig. 9. Data showed that the temperature coefficient of PVG dosifilm from  $20^\circ\text{C}$  to  $55^\circ\text{C}$  was approximately  $+0.008$  of the response/ $^\circ\text{C}$ .

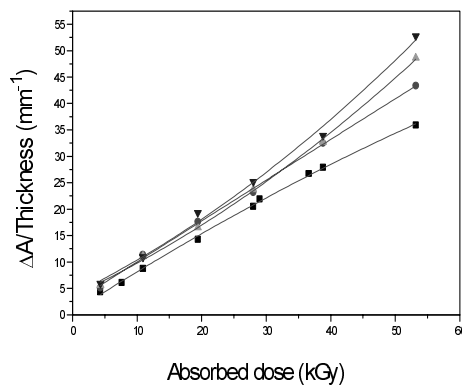


FIG. 8.  $\Delta A/\text{Thickness} (\text{mm}^{-1})$  vs. absorbed dose at different irradiation temperatures.  $\blacksquare$   $-20^\circ\text{C}$   $\bullet$   $-35^\circ\text{C}$   $\blacktriangle$   $-45^\circ\text{C}$   $\blacktriangledown$   $-55^\circ\text{C}$ .

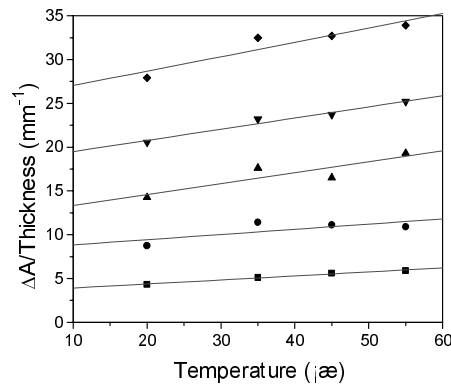


FIG. 9.  $\Delta A/\text{Thickness} (\text{mm}^{-1})$  vs. irradiation temperature at various absorbed doses.  $\blacksquare$   $-4.3 \text{ kGy}$   $\bullet$   $-11 \text{ kGy}$   $\blacktriangle$   $-19.5 \text{ kGy}$   $\blacktriangledown$   $-29 \text{ kGy}$   $\blacklozenge$   $-39 \text{ kGy}$ .

Thus, the value of the specific net absorption at any temperature can be expressed as follows:

$$\Delta A/\text{Thickness}_{(t^\circ\text{C})} (\text{mm}^{-1}) = \Delta A/\text{Thickness}_{(20^\circ\text{C})} (\text{mm}^{-1}) [1 + (t^\circ\text{C} - 20) \times 0.008] \quad (5)$$

where, the subscripts ( $t^\circ\text{C}$ ) and ( $20^\circ\text{C}$ ) refer to the temperature during irradiation.

The experimental values (<sup>1</sup>Exp.) and the calculated values (<sup>2</sup>Cal or <sup>3</sup>data) of  $\Delta A/\text{Thickness}_{(t^{\circ}\text{C})}$  ( $\text{mm}^{-1}$ ) for PVG dosifilm (i.e. calculated values from Eq. (5) and Eq. (2)) are listed in Table II. At 20°C, the deviation between <sup>1</sup>Exp and <sup>3</sup>data is about  $\pm 2\%$ . This reflects the degree of goodness of fit for regression of Eq. (2). The deviation between <sup>1</sup>Exp and <sup>2</sup>Cal values was about  $\pm 5\%$ . This reflects the experimental temperature correction. Total deviation was less than 10%, i.e. less than the uncertainty of the routine dosimetry system. Thus, Eqs (2) and (5) can be used to determine absorbed dose at any irradiation temperature between 20 and 50°C.

TABLE II. EXPERIMENTAL VALUE  $\Delta A/\text{THICKNESS}_{(t^{\circ}\text{C})}$  ( $\text{mm}^{-1}$ ) AND CALCULATED VALUE BY EQ. (5) OF PVG DOSIFILM AT VARIOUS TEMPERATURES.

Irradiation temperature (°C)	10.9 kGy		19.4 kGy		28 kGy		39 kGy	
	$\Delta A/\text{Thickness}$ ( $\text{mm}^{-1}$ )		$\Delta A/\text{Thickness}$ ( $\text{mm}^{-1}$ )		$\Delta A/\text{Thickness}$ ( $\text{mm}^{-1}$ )		$\Delta A/\text{Thickness}$ ( $\text{mm}^{-1}$ )	
	<sup>1</sup> Exp.	<sup>2</sup> Cal.	<sup>1</sup> Exp.	<sup>2</sup> Cal.	<sup>1</sup> Exp.	<sup>2</sup> Cal.	<sup>1</sup> Exp.	<sup>2</sup> Cal.
20	8.75	<sup>3</sup> 8.82	14.26	<sup>3</sup> 14.92	20.54	<sup>3</sup> 20.83	27.92	<sup>3</sup> 27.42
35	11.4	10.06	17.6	16.4	23.2	23.62	32.5	32.11
45	11.1	10.94	16.5	17.82	23.7	25.67	32.7	34.9
55	10.9	11.81	19.3	19.25	25.2	27.73	33.9	37.6

<sup>1</sup>Exp.- experimental values,

<sup>2</sup>Cal. — the values calculated by Eq. (5),

<sup>3</sup>data — values calculated by Eq. (2).

### 3.2.4. Effect of relative humidity on PVG dosifilm for EB irradiation

The relationship of  $\Delta A/\text{Thickness}$  ( $\text{mm}^{-1}$ ) vs. dose at various relative humidity for irradiated PVG dosifilm is shown in Fig. 10. Figure 11 indicates that at the same dose specific net absorption increases with increasing relative humidity from 0% to 75.7%, with humidity coefficient of about 0.006 of the response/% r.h. The change in the specific net absorption of the irradiated PVG dosifilm for various doses from 0% to 33% r.h. was less than 4%, therefore the effect of relative humidity on irradiated PVG dosifilm could be neglected below 33% r.h. However, at higher relative humidity, the effect on PVG dosifilm irradiated by EB can not be neglected, i.e. the value of the specific net absorption must be corrected by the coefficient 0.006/% r.h.

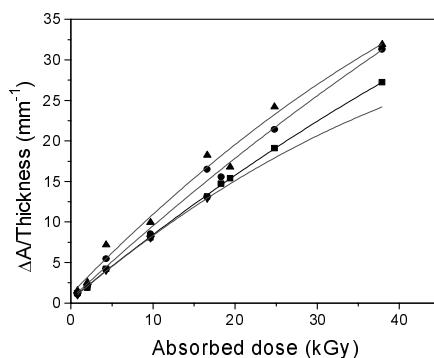


FIG. 10. Specific net absorbance vs. absorbed dose at various relative humidity.  $\blacktriangledown$  — 0% r.h.  $\blacksquare$  — 33.6% r.h.  $\bullet$  — 54.9% r.h.  $\blacktriangle$  — 75.7% r.h.



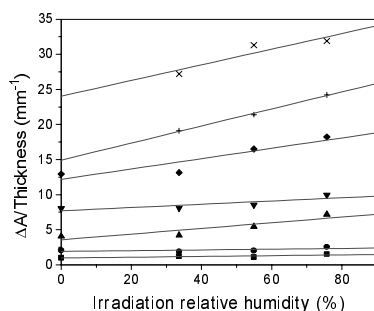


FIG. 11. Specific net absorbance vs. relative humidity at various absorbed doses. ■  $-0.8$  kGy  
●  $-2$  kGy ▲  $-4$  kGy ▼  $-10$  kGy ◆  $-17$  kGy ✱  $-38$  kGy.

### 3.2.5. Effect of dye and additive on dosimetric characterization of PVG dosifilm

The concentration of the dye and additive affected the dosimetric characterization of PVG dosifilm irradiated by gamma rays. The dose response sensitivity ( $\Delta A/\text{Thickness}$  ( $\text{mm}^{-1}$ ) vs. dose) increased from 0.102 to 1.522 with the increase of LMG concentration from  $10^{-5}$  to  $10^{-4}$  ( $\text{mol}/\text{g}_{\text{PVB}}$ ) in dose range from 1 kGy to 44 kGy (see Fig. 12). The dose response sensitivity increased from 0.711 to 2.313 with the increase of the ratio LMG/RX from 0.5:1 to 2:1 (see Fig. 13). Effects of the concentration of the dye and additive on PVG dosifilm irradiated by EB are currently under investigation.

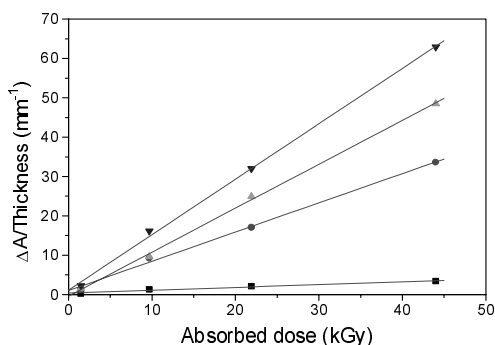


FIG. 12. Effect of LMG concentration on dose response sensitivity. LMG concentration: ■  $1 \times 10^{-5}$  mol/g<sub>PVB</sub> ●  $5 \times 10^{-5}$  mol/g<sub>PVB</sub> ▲  $8 \times 10^{-5}$  mol/g<sub>PVB</sub> ▼  $1 \times 10^{-4}$  mol/g<sub>PVB</sub>.

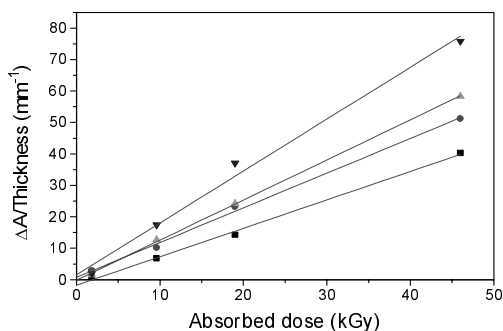


FIG. 13 Effect of additive concentration on dose response sensitivity. LMG/RX: ■  $-0.5:1$  ●  $-1:1$   
▲  $-1.5:1$  ▼  $-2:1$ .

### 3.3. Stability and reproducibility of PVG dosifilm

#### 3.3.1. Influence of storage condition on PVG dosifilm

The PVG dosifilm is slightly sensitized by sunlight. Figure 14 shows the change in absorbance of unirradiated PVG dosifilm stored in dark and in room light. The change in  $A/A_0$  was less than 3% in dark for more than two months, and was more than 5% in room light. However, the absorbance changes significantly under sunlight. Thus, it is possible to operate in room light, but it is preferred to store it in dark. Also, the absorbance increased by about 20% when stored at 60°C for 24 h. Irradiated PVG dosifilm was stable for long term storage at room temperature; the increase in absorbance was less than  $\pm 3\%$  over a period of more than two months.

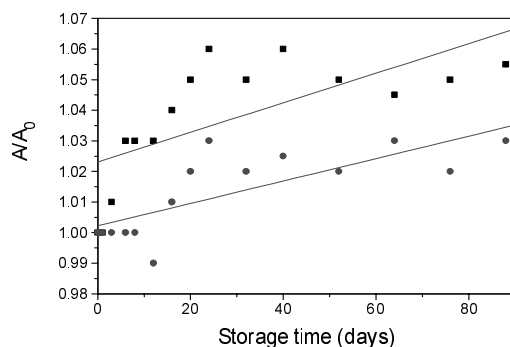


FIG.14. Effect of storage condition on unirradiated PVG dosifilm. • — stored at room temperature in dark, ◻ — stored at room temperature in room light.

#### 3.3.2. Measurement dispersion of PVG dosifilm

A set of five PVG dosifilms was irradiated by gamma rays and also by EB. The measurement readout dispersion were less than  $\pm 4\%$  in the absorbed dose range 5 to 140 kGy. The dosimetric response of PVG dosifilm for every batch may be different. Therefore, calibration is necessary for each batch. The dose response sensitivity for batches from J-11 to J-24 was  $1.065 \pm 0.044$  and  $0.783 \pm 0.031$  for gamma rays and EB, respectively, for the dose range 5 to 100 kGy; thus, batch-to-batch variation is about 4%.

## 4. CONCLUSION

Experimental results have shown that the PVG dosifilm can be used as a routine dosimeter for gamma rays and for EB irradiation. The dosimetric sensitivity (at 627 nm) of PVG dosifilm irradiated by gamma rays was  $1.065 \pm 0.044$  in the dose range from 5 to 100 kGy, and for EB it was  $0.783 \pm 0.031$  in the dose range from 5 to 140 kGy. The relationships of specific net absorbance vs. absorbed dose was found to be sub-linear. The specific net absorbance of irradiated PVG dosifilm was affected by irradiation temperature and relative humidity. The average value of the irradiation temperature coefficient was  $0.008/^\circ\text{C}$  in the temperature range  $20^\circ\text{C}$  to  $55^\circ\text{C}$ . Relative humidity effect on irradiated PVG dosifilm can be neglected below 30% r.h., but the correction was necessary at higher relative humidity by the average value of the coefficient  $0.006/\%$  r.h.. The irradiated PVG dosifilm must be stored for at least 24 h after irradiation before absorbance measurement due to the post-irradiation effect. The total uncertainty in the dose measured by PVG dosifilm is less than 10% for EB irradiation.

# CALORIMETRY FOR ABSORBED DOSE MEASUREMENT AT 1–4 MeV ELECTRON ACCELERATORS

A. MILLER  
Risø High Dose Reference Laboratory,  
Risø National Laboratory,  
Roskilde, Denmark

## Abstract

Calorimeters are used for dose measurement, calibration and intercomparisons at industrial electron accelerators, and their use at 10 MeV electron accelerators is well documented. The work under this research agreement concerns development of calorimeters for use at electron accelerators with energies in the range of 2–4 MeV. The dose range of the calorimeters is 3–40 kGy, and their temperature stability after irradiation was found to be sufficient for practical use in an industrial environment. Measurement uncertainties were determined to be 5% at  $k = 2$ .

## 1. INTRODUCTION

A polystyrene calorimeter for use at 10 MeV electron accelerators consists of a polystyrene disk situated within thermally insulating polystyrene foam [1]. The disk is 18-mm thick, with a diameter of 138 mm and the foam thickness is about 50 mm. The temperature is measured with a calibrated thermistor that is inserted into the side of the polystyrene disk. At this thickness the dose distribution through the disk is almost a straight line.

The temperature is measured before and after irradiation, and the temperature increase  $\Delta T$  is in principle related to absorbed dose  $D$  by the relationship:

$$D = \Delta T * C_p \quad (1)$$

where,  $C_p$  is the specific heat of polystyrene. This value is, however, not well known and instead a calibration is established by comparison of the temperature rise ( $\Delta T$ ) with the absorbed dose as measured with reference dosimeters. The reference dosimeters (usually alanine) are irradiated in the centre of an absorber with identical geometry to that of the calorimeter. Thus the dose to the reference dosimeter equals the average dose to the calorimeter. The relationship between dose and temperature becomes of the form:

$$D = \Delta T * (K_1 + T_{ave} * K_2) \quad (2)$$

where,  $T_{ave}$  is the average temperature of the polystyrene disk, because the specific heat is a function of temperature.

## 2. DEVELOPMENT OF THIN CALORIMETERS

Calorimeters with 18 mm thickness are not useful at energies of 2 MeV. At this thickness the calorimeter becomes totally absorbing, and although a temperature increase can be measured after irradiation, it is not easy to relate this to absorbed dose through calibration as described above. Thinner absorbing disks have to be employed, and for use at 2 MeV a disk thickness of 2 mm was tested with a very small thermistor for measuring temperature. However, the radiation induced temperature of this disk dropped too rapidly after irradiation,

and instead a sandwich of three 2-mm disks was tested, with the temperature being measured only in the middle disk. The disks were not thermally insulated from each other, but we observed that there was only a very slow rate of heat exchange between them.

Initial testing of the 2 MeV calorimeter was done at the Risø 10 MeV electron accelerator, where the disks were placed in the normal foam insulation with a total thickness of 100 mm, but height restrictions at the 2 MeV electron accelerator, where real life testing was to be carried out, made it necessary to design a new foam insulation with a total thickness of only 28 mm. This thin insulation made the cooling faster than with the 100 mm insulation, approximately 2.5% per minute after irradiation. However, under reproducible irradiation conditions this cooling could be corrected for, and it did not add significantly to the overall measurement uncertainty.

Calibration of this calorimeter could be carried out by using an absorber that would place alanine dosimeters at the same position as the middle of the three disks giving the same type of calibration relationship as in Eq. 2. In the same way the calorimeter could be used for calibration of thin film dosimeters when these were placed between two 3-mm plates located in the thermal insulation. That would ensure that the dose to the film dosimeters was the same as that to the calorimeter.

The lowest dose that could be measured by the calorimeter was determined by the smallest temperature increase that could be measured reproducibly. This was approximately 2°C corresponding to 3 kGy. The largest dose was determined by the maximum temperature at which the thermistors were calibrated, 55°C, leading to a maximum dose of 40 kGy for 25°C environmental temperature.

The components of the measurement uncertainty are:

Calibration by alanine reference dosimeters (type B):	1.5 %
Measurement of temperature (type A):	1.0 %
Establishment of calibration function (type A):	1.0 %
<u>Difference in irradiation geometries (type B):</u>	<u>1.5 %</u>
Total overall uncertainty:	2.5 %
At k = 2:	5.0 %

### 3. CONCLUSION

The 2 MeV calorimeters are now in use for documentation of radiation sterilization at one industrial electron accelerator and are being tested at two others.

### REFERENCE

- [1] AMERICAN SOCIETY FOR TESTING AND MATERIALS, Practice for use of calorimetric dosimetry systems for electron beam dose measurements and dosimeter calibrations, ASTM E1631, Annual Book of ASTM Standards, 12.02, ASTM, 1998

# **MODELIZATION OF THE PHYSICAL AND CHEMICAL PHENOMENON OF FADING AND CONTROL OF PARAMETERS FOR QUALITY ASSURANCE OF ESR/ALANINE MEASUREMENTS**

J.M. DOLO, V. FEAUGAS, E. PICHOT

Commissariat à l'énergie atomique,

Département des applications et de la métrologie des rayonnements ionisants,

Gif-sur-Yvette, France

## **Abstract**

The work of L.M.R.I. according to the research agreement contract is constituted of three parts which all contributed to the same objective: the better understanding of the phenomenon of production and disappearing of free radicals in the alanine dosimeter. The first part is the contribution to the accuracy of the measurements parameters and their optimisation. Many parameters such as the power level and the modulation amplitude must be chosen carefully. This part describes how we have defined the limits of the parameter values giving the best arrangement between the four objectives: the better signal-to-noise ratio with the highest amplitude of the central line, and the better reproducibility with a resolution as good as possible. The second part is our contribution to increase the understanding on how external parameters influence the chemical reactions in alanine. Two experimental studies of combined effects have been realised in the laboratory. A multi-parametric approach (experimental design) has shown the complexity of the problem. The chosen methodology permits to give the first global quantitative view of the fading. However the obtained results, that suggested a physico-chemical recombination reaction of the free radicals created during the irradiation, are not sufficient. The last part is a study of the kinetic aspect of the mechanism. The aim of this study is to indicate the number of transient species leading to this radical and their kinetics of reaction. We follow the evolution of the ESR/alanine spectrum shape and correlate the response estimated from the central peak height to the absorbed dose. From these hypothesis we modelize the kinetics of free radical densities and check their fitting with experiment.

## **1. INTRODUCTION**

The idea to use alanine for dosimetry for evaluation of dose originated in the sixties [1], but its development for applied metrology in industrial applications: food irradiation, radiation processing for polymers has been successful for high doses (1–100 kGy). Since several years, with the increasing quality of ESR spectrometers, the measurements of absorbed doses in water for radiotherapy treatments or blood sterilisation (1–100 Gy) seems to be accessible to alanine dosimetry [2, 3]. The non-destructive measurement, and some other advantages have given to this method many attraction for use as routine dosimetry method for the quality control of the radiation processing with a sufficient level of guarantee. But it is hoped that this method is also a good transfer dosimetry system reliable and traceable to a primary standard for international comparisons with an accurate level for the mastery of the uncertainties under a quality assurance system. Some laboratories, have realised important progress in this domain. Nevertheless, the most important difficulty for using ESR/alanine as a measurement control system for comparisons is to characterise, to evaluate and to quantify the influence of physic and chemical factors on the evolution of the ESR signal of an alanine dosimeter over a sufficient time period.

## **2. CONTRIBUTION TO ACCURACY OF MEASUREMENT PARAMETERS**

In ESR/alanine dosimetry, the absorbed dose of an alanine sample is determined through its absorption spectrum. The recording of an ESR spectrum requires selection of the values of several parameters of the spectrometer. First we have to choose the interval of the magnetic field intensity in which the spectrum will be recorded. We have stated the sweep

width, the intensity of the magnetic field at the middle of the spectrum and the intensity of the magnetic field at the beginning of the spectrum. The microwave attenuation, the gain, the amplitude modulation, the phase modulation, the time constant, the sweep rate, the resolution in X and the number of scans have also to be set. The number of parameters show that the problem is complex and that a monoparametric study would not probably give satisfactory results. The choice of the parameters is strongly linked with the qualities of the spectrum that we attempt to obtain. We have chosen to select the value of the measurement parameters in order to get a sufficient reproducibility in the spectrum to be sure that we can get from it a valid measure (representative of the number of radicals) and if we observe a variation between two spectra, this deviation is significant. This subject has been approached by different ways. First, a bibliographic study was carried out in order to compare the different choices in the setting of parameters and the reasons given by the authors. Then, several experimental designs were realised without taking into account the values found in the literature.

We have selected some articles dealing with ESR dosimetry in which the given information about the experimental conditions was sufficient to allow us to perform the experiments in our laboratory. The materials used by the authors of these articles are listed in Table I [4–10]. The analyses will state three categories according to the dose range of work: less than 100 Gy [5, 6, 8], around 100 Gy [4–7, 10] and in the kilogray range [4, 9, 10]. The measurement parameters of these laboratories are presented in Table II. A safe rule of choice is to make sure that the time needed to scan through the ESR spectrum is ten times greater than the length of the time constant [11, 12] for precise dosimetry to record an ESR spectrum as undistorted as possible. Therefore, the time constant should be about 1/40 of the scanning time of the central peak. The value of this parameter is linked to the sweep rate. The receiver gain will depend on the dose delivered to the sample. It must be high enough to get a good signal-to-noise ratio, but not too high to saturate the signal [11]. Signal averaging involves repeatedly acquiring the spectrum and adding them together. The signal increase is proportional to the number of scans (N), whereas the noise increase is proportional to  $\sqrt{N}$ . The signal-to-noise ratio is then proportional to  $\sqrt{N}$  [11]. The microwave power of our

TABLE I. MATERIALS USED BY DIFFERENT LABORATORIES

Laboratory	Spectrometer	Dosimeter
Risø (Miller 1996) [4]	Bruker EMS104	pellet: 85% L- $\alpha$ -alanine, 15% paraffin wax
ISS (Bartolotta 1993) [5]	Bruker ESP300 TE102 cavity	pellet: 80% of polycrystalline L-alanine, 20% paraffin
AERIAL (Kuntz 1996) [6]	Bruker ESC106 9105 TMH280 cavity	pellet: L- $\alpha$ -alanine, binding (3%), lubricating agent (2%)
NIST (Nichiporov 1995) [7]	Bruker ESP300E and ECS 106 TMH cavity	pellet: 90% L- $\alpha$ -alanine, polyethylene
NPL (Sharpe 1996) [8]	Bruker ESP300	pellet: 90% L- $\alpha$ -alanine, 10% paraffin wax
GSF (Wieser 1989) [9]	Varian E-9	pellet: 80% of polycrystalline L-alanine, 20% paraffin
JAERI (Kojima 1986) [10]	JEOL FE3X	film: polymer-alanine — 33.3/66.7 by wt % polymer/ethylene-propylene

TABLE II. INSTRUMENT PARAMETERS CHOSEN BY DIFFERENT LABORATORIES TO RECORD THE EPR SPECTRUM OF IRRADIATED ALANINE

Laboratory	Microwave power (mW)	Modulation (mT)	Sweep width (mT)	Sweep time (s)	Sweep rate (s/mT)	Time constant (ms)	Number of sweeps	Range of dose	Reasons of these choices
Riso (Miller 1996) [4]	1.25 1.25	0.2 0.2	2 2	10 20	5 10	20 320	4 10	kGy 100 Gy	no information
ISS (Bartolotta 1993) [5] AERIAL (Kuntz 1996) [6]	1 8	1 1	2 2	21	24 10.5	328	1 5	0.5-100 Gy 3.6-120 Gy	best signal to noise ratio even though a line distortion can be produced no information
NIST (Nichiporov 1995) [7]	10	1	8	41	5	655	1	10-300 Gy	no information
NPL (Sharpe 1996) [8]	6	0.6	20	20	1	163	3	below 10 Gy	maximum signal to noise ration consistent with the preservation of the major features of the alanine spectra no information
GSF (Wieser 1989) [9]	1	0.1	2.5	84	33.6	640	1	1 kGy-50 kGy	reproducibility of the peak to peak amplitude despite modulation broadening and power saturation
JAERI (Kojima 1986) [10]	4	1	20	240	0.08	30	1	100 Gy-30 kGy	

spectrometer can vary between 200 mW and 0.2 mW. The amplitude of the observed ESR signal is proportional to the polarisation of the paramagnetic species. The system is called saturated when the transitions rates towards the high and the low levels are equal. There is no more energy transfer between the microwave and the spin system. When saturation is reached, the area under the absorption curve is no more a valid measure of the spin concentration. For a given power, it depends on the sweep rate, the amplitude modulation and the frequency [13]. Saturation occurs when the intensity of the signal according to the microwave power is no more linear [14]. In the case of alanine, the saturating power values found in the literature are between 0.3 mW and 10 mW. According to Wieser [14], if the modulation amplitude is high, the satellite lines are not visible and saturation only occurs around 10 mW. If the modulation amplitude is lower than the line width (0.5 mT), then saturation occurs at about 0.3 mW. Saturation curves have been obtained by Wielpolski [15] for both transitions using the height of the signal over the base line instead of the peak to peak amplitude. With a modulation amplitude of 0.05 mT, the central peak saturates at about 5 mW, the satellite at about 17 mW.

According to Weil [13], the optimum setting of the modulation amplitude depends on how much sensitivity can be sacrificed for faithful reproduction of the line shape or vice versa. A reasonable compromise between these two objectives is achieved by reducing the modulation amplitude by a factor of 4–5 from the value that makes the signal amplitude a maximum. When the modulation amplitude is low enough, one can observe the central peak of the alanine spectrum as well as the satellites lines. Arber [16] have studied the spectrum of irradiated alanine in X band according to the microwave power and the frequency. The central peak is accompanied on either side by a pair of satellite lines each separated from the main peak by ca 0.5 mT. These lines have been attributed by Van Laere [17] to secondary species despite the fact that they have never been observed. A stable radical would dominate at low microwave power (~1 mW) whereas at high microwave (~100 mW), the spectral characteristics of the stable radical are saturated and new ones can be detected. Rakvin [18] considers several hypothesis. One is the existence of two forms of stable radical. The second is that there are two radicals with different rotation speeds of the methyl group. This phenomenon may also be explained by the existence of a stable radical and a transient species. Arber [16] attribute these satellite lines to spin-flips. Spin-flips are forbidden transitions involving a simultaneous change in the electron spin quantum number and the proton spin quantum number of the methyl protons of the neighbouring molecules.

For our experimental part, we have worked with pure L- $\alpha$ -alanine powder (grain size between 150 and 180  $\mu\text{m}$ ) supplied by Merck, inserted in suprasil tubes. Our spectrometer is the X band EMX model of Bruker, with a TMH 4108 cavity. The value of the magnetic field intensity at the middle of the spectrum is set by the content of the cavity: the sample, the tube and the temperature controller. This value is close to 0.3485 T without the temperature controller, and 0.3410 T with it. The sweep width is 15 mT in order to record the total spectrum. We have recorded spectra (see Fig. 1) at three different doses: 5 kGy, 100 Gy, 5 Gy with the measurement parameters listed in Table II. Comparing these spectra, we notice that they have not all the same shape. Time constant filters out noise by slowing down the response time of the spectrometer. As the time constant is increased, the noise level will drop. If the chosen time constant is too long for the rate at which the magnetic field is scanned, the signal can be distorted or even filtered. To obtain the highest ESR signal amplitude, we have recorded ESR spectra at attenuation varying between 16 and 24 dB (microwave power between 0.79 and 5.02 mW) and the modulation amplitude between 0.05 and 0.6 mT. The significant effects on the signal amplitude are, in order, due to the modulation amplitude, the



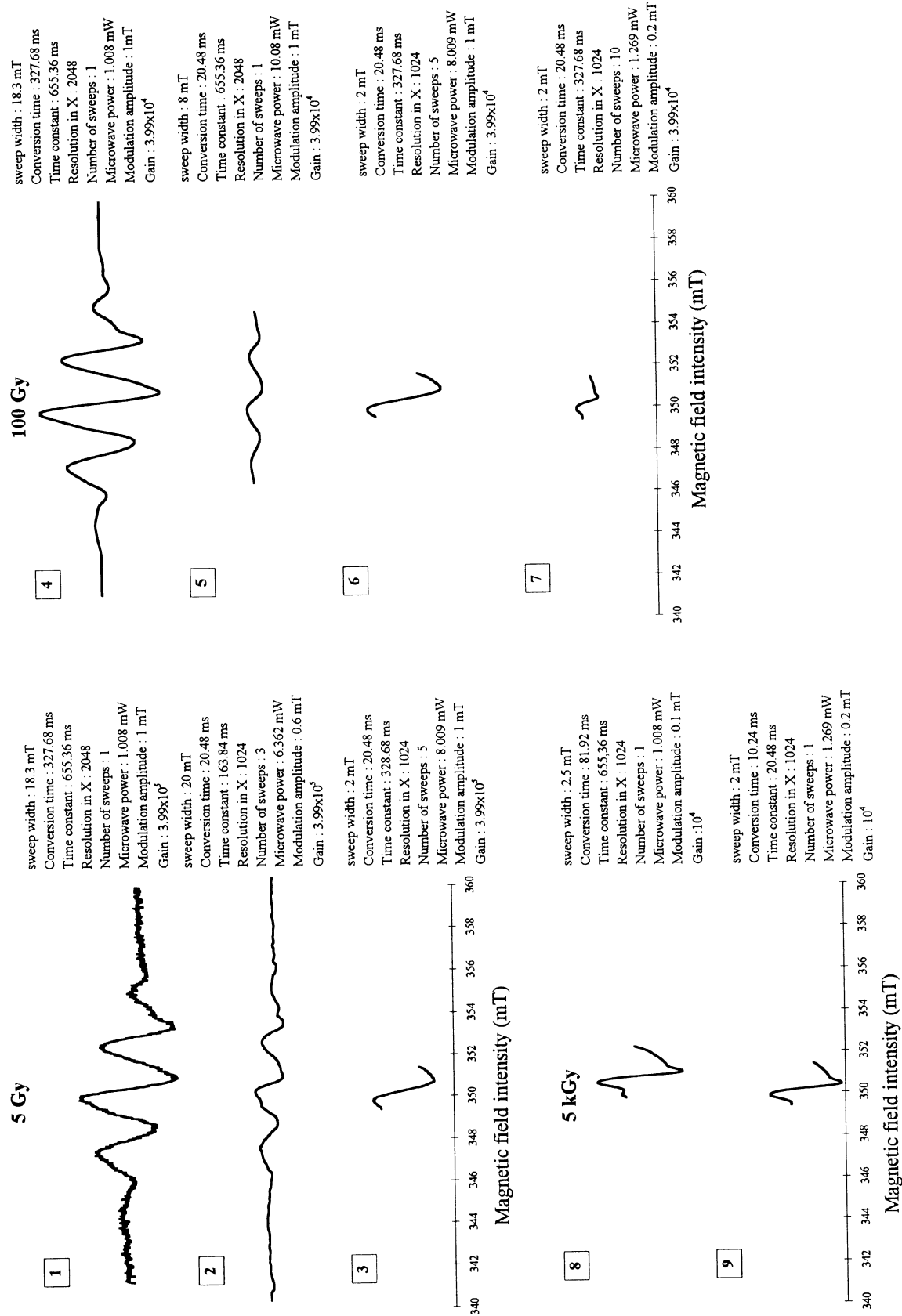


FIG. 1. Spectra recorded with the measurement parameters listed in Table II

attenuation, their coupling, and modulation square effect. The mathematical model explains 99.5% of the observed variability in the signal amplitude. An increase of the modulation amplitude has a stronger positive effect on the amplitude for low attenuation than for high one. An increase of attenuation has a stronger negative effect on amplitude for a high modulation amplitude than for a low one. Another experimental design has been realised with two factors: attenuation and sweep time. Attenuation was set between 12 and 22 dB, sweep time between 10.49 and 335.54 s. The significant effects on the signal amplitude are, in order, due to the sweep time, then the attenuation, then their coupling and finally the squared of the attenuation. The increase of sweep time has a greater positive effect on the amplitude when attenuation is low. If sweep time is low, a decrease of attenuation has only a slight effect on the value of the amplitude. The obtained model explains 99.8% of the observed variability in the amplitude. Our objective is not only to have the highest amplitude but also to obtain an undistorted and unsaturated spectrum. The increase in the modulation amplitude has for consequence an increase of the amplitude of the ESR signal and a broadening of the lines. For a 0.3 mT modulation, the increase of the line width compared to the width at 0.05 mT is greater than 3.5%, independent of the value of the power (between 0.796 and 5.024 mW). For a 0.2 mT modulation, it is less than 3%. The modulation amplitude should be 0.25 mT in order to limit the lines broadening. To determine the saturating power, the curve representing the ESR signal amplitude vs. the square root of the microwave power has been drawn for a modulation amplitude of 0.25 mT. Saturation occurs when the amplitude is no more linear. The saturating power for an amplitude modulation of 0.25 mT is 1.26 mW. The influence of sweep rate on the value of the saturating power has been studied for sweep rates varying between  $5.12 \times 10^{-3}$  and 0.163 s/channel. To record several spectra for a sample (because of the anisotropic response when the sample is rotated), the maximum duration of the recording of a spectrum has been limited to 335.54 s. No influence of the sweep rate on the value of the saturating microwave power has been observed. The choice of sweep time and time constant will depend slightly on the number of spectra necessary to ensure a good repeatability and reproducibility of measurement despite the problem of anisotropic response of the sample.

The choice of the measurement parameters depends on the objectives one wants to achieve and on the dose received by the sample. As we measure the response to the dose on the central peak, the sweep width is 2 mT. As we are working in the kilogray range, the signal to noise ratio is high. Consequently, recording several scans is pointless. But as shown before, in the gray range, signal averaging is very useful. We found that recording the alanine spectra at a microwave power of 1.260 mW and an amplitude modulation of 0.25 mT enables to get an undistorted and an unsaturated signal.

### 3. DEVELOPMENT OF OUR EXPERIMENTAL DESIGNS

We have chosen to use the method of experimental design for two reasons. The experimental factors are numerous and their natures are very different. They have probably combined effects. This methodology enables us to choose the most important controlled parameters as factors. We can observe simultaneously their influence on the response and quantify it on a large domain and limit the number of experimentation. The physical/chemical nature of the dosimeter, the relative humidity rates before and after irradiation, the dose rate and the dose are the chosen factors. The storage duration after irradiation is not exactly a controlled factor but it is integrated the same way as in the array of measurements since we study the evolution of the dosimeter signal with time. We have chosen to work on three types of alanine probes: pure alanine, the blend not compacted and compacted in a pellet. Pure

alanine will be our reference and the pellet the object of our study, the comparison being used to highlight some possible phenomena of diffusion. This controlled factor is discontinuous, so no interpolation is allowed between the different states of alanine. The influence of the relative humidity before and after irradiation has been studied for many years [1, 19], water is probably the main cause of the recombination of free radicals. In order to understand the influence of water on the stability of the free radicals of alanine, that is to say the role of water in the mechanism of recombination, we have chosen to control four humidity levels before and after irradiation: 0, 20, 40, 60%, temperature being maintained by air conditioning to room temperature (about 20°C) [20]. The balance is obtained for a contact of five weeks at least. The storage after irradiation is systematic all along the experience. These two factors are continuous, so that the results could be interpolated among 0 and 60% relative humidity. The influence of the dose rate on the signal evolution has been studied [21]. Nevertheless, we wish to control it in order to test its influence mainly during the first days. We have used two dose rates: 4 kGy/h and 300 Gy/h. This parameter is continuous so interpolation is allowed. The importance of dose on the signal amplitude is obvious. We have chosen to work on doses between 100 Gy and 10 kGy. This factor is continuous and we wish to be able to interpolate the best as possible within the chosen area. We have followed more particularly two areas of time: the first ten days and the period between the tenth and the fortieth day. Different arrays of experience have been elaborated so. One physical state and one dose rate are attributed to each array. We have realised in each case three series of arrays of experiences, from 1 to 3 days, from 4 to 10 days and from 17 to 39 days. Details on the factors are listed in Table III. The results have been exploited by the use of a dedicated software. A purely mathematical approach of the results enables to distinguish the influence of each factor and to quantify it. For the handling with alanine powder, we have attributed a tube to each experience with mass of about 50 mg. The tube is positioned in a reproducible manner in the ESR cavity with the greatest care. The adjustment of the spectrometer is mainly subjected to the measurement of the lowest dose. We have realised the spectra recording in the following conditions: 4 scans of 41 seconds on 2048 channels, a field sweep width of 100 Gauss, a field modulation amplitude of 1 Gauss, a time constant of 328 ms with a power of 10 mW.

TABLE III. CONTROLLED FACTORS

Factor	Nature of factors	Area of control	Suggested values
Dose	continuous	1 Gy –50 kGy	100, 300, 500, 1000, 5000, 10000 Gy
Dose rate	continuous	200 Gy/h–4 kGy/h	200 Gy/h and 4 kGy/h
Humidity before irradiation	continuous	0–60%	0, 20, 40, 60%
Humidity after irradiation	continuous	0–60%	0, 20, 40, 60%
Alanine forms	discontinuous		pure alanine, LMRI mixture, pellet
Time after irradiation	continuous	0–40 days	1, 2, 3–4, 7, 10, 17, 28, 39

The analysis is done in two stages: the first gives a general view of the measurements, while the second gives a more precise view highlighting the evolution noticed during the first days of the experience. The first finding showed the overwhelming influence of the dose factor; it was expected and was not the major researched result of our study. The chosen values for the dose rate have implied variation in dose for an easy management of irradiation duration. Nevertheless, it should be specified that the structure of the array of experiences is a function of the influence attributed to the controlled factors. The importance of this factor do not give us an optimum exploitation of the array of experience and so we have only done graphical exploitation. To observe the effects of the other controlled factors, we have used two types of graphic of the results for our analysis. First we have drawn surfaces of the results according to time and humidity before or after irradiation, while other factors were fixed (Figs 2a, 2b and 2c). Then we have exploited the iso-signal curves according to humidity with a constant delay after irradiation (Figs 2d, 2e and 2f). The first solution offers a total view of the signal evolution with time, the second permits a detailed day-to-day analysis; this is possible due to a more precise visualisation of the height values.

A quick study of the surfaces of the signal evolution for doses from 1 to 10 kGy shows two essential things. The behaviour of alanine is the same however its form is: pure or pellets. Under some conditions, the amplitude of the signal is clearly more important since the first days of measurements, then it decreases proportionally to humidity after irradiation, independent of the storage conditions before irradiation. The humidity before irradiation seems to result in a variation in the dosimeter response, characterised by a hump of amplitude for a storage after irradiation at low humidity rate and by an increase ending in stage for a storage at 60%. This finding is true for doses from 1 to 10 kGy. His importance is about 5% for the doses from 5 and 10 kGy and reaches 15% and 10% respectively for alanine and pellets at 1 kGy. The moisture after irradiation takes part in the hump phenomenon before the first day of measurement. But its main action, already observed [21], revealed itself with time according to its importance. If the moisture after irradiation is set to 60%, one observes a decrease of the amplitude of about 5% for doses of 5 and 10 kGy, of 20% for doses of 1 kGy, almost independent of the storage conditions before irradiation. The previous findings show the influence of the dose on the signal evolution. An increase of the dose leads to two main changes on the signal evolution, it cancels progressively the total phenomenon of increase for relative humidity of 0% after irradiation, and favours the decrease of the amplitude for storage conditions of 60% after irradiation. By studying the surfaces of the results for doses from 1 to 10 kGy, one observes both the importance of time that reveals the effects of the other factors and the weak influence of the controlled factor time strictly speaking. The decrease of the signal is a combined phenomenon, but the main contribution comes from relative humidity after irradiation. However, the study of these surfaces for doses of 100, 300 and 500 Gy shows the lack of amplitude variation against the moisture levels. The signal decreases by 5% with time. The interpretation of this finding is complex, because if the doses are lower than before, the dose rate is also much lower. It is impossible to come to a conclusion about the influence of these factors in this case. The evolution is inversely proportional to the relative humidity after irradiation. This confirms the role of water on the recombination reactions of the alanine free radical. The total increase of the amplitudes established for a 0% relative humidity suggests the creation with time of these same free radicals. The mechanism of formation can be explained by the presence of other free radicals coming from alanine [22], although these are not already identified. These radicals would progressively change into the "classical" free radicals. The increase of the amplitude with humidity after irradiation, for high conditions of humidity before irradiation, can be explained the same way if we suppose that a certain type

of free radical reacts quickly in the presence of water. The hump of amplitude probably comes from a balance in water inside the dosimeter before irradiation.

For the analysis of the results with the help of iso-signal curves, we have fully used the possibilities of the arrays of experiences that quantify the influence of controlled factors on the three areas of measurements. The study of the results of the different arrays shows that the influence of the moisture before irradiation is limited to the first days, and later the influencing factor is moisture after irradiation. One observes also since the first day a change of amplitude according to the storage conditions before irradiation. Finally, an area of perturbation of measurements between the second and the fourth day is observed for all the experimental conditions used: a decrease of about 2% of the signal, and it being the only area having a so remarkable dispersion of the results. The amplitude of the signal during the first days is function of the storage conditions before and after irradiation. Whatever the physical state of the probe is, the evolution of the signal during the first three days can be summed up by an increase of the signal for relative humidity after irradiation of 0%, and by a decrease of signal proportional to the relative humidity since 20 to 30%. These two effects lead to a regrouping of the response amplitudes. In the case of pure alanine, one clearly observes since the fourth day the influence of the humidity after irradiation. It is also the case for the pellets, but in a more progressive manner between the fourth and the tenth day. The evolution of the curves between the tenth and the fortieth day highlights a certain stability for the storage conditions before irradiation of 30% r.h., and the more important decrease of the signal in the presence of a high relative humidity. It is noticeable on the curves coming from the last experiences array that a rapid humidity change is harmful for the signal stability. The analysis of the iso-signal curves confirms the observed trends with the response surface, and they also give some new information. The most important perturbations observed between the second and the third day need some confirmation by other measurements, but this observation is noted in an article [22]. The difference between the amplitudes, since the first day, among 0 and 60% r.h. before irradiation is confirmed, but the existence of a hump in the amplitude for storage before irradiation at 20 to 30% r.h. is not visible before the 17<sup>th</sup> day. Its existence in the first days must be confirmed. A study on the importance of the parameters with the help of the three matrices of experimentation show that pure alanine is more rapidly sensitive to relative humidity after irradiation than the pellets. This observation is explained by the difference between the diffusion speed of water molecules with the compactness of the material.

#### 4. KINETICS APPROACH

In ESR/alanine dosimetry, many researchers make a general assumption that the ESR signal is only due to a stable free radical. But in fact, the mechanism of radical formation in a powder is not well determined. The first step of this understanding is in the definition of the organisation of the crystal at the beginning. L- $\alpha$ -alanine form has the formula  $\text{CH}_3\text{CH}(\text{NH}_3)^+\text{CO}_2^-$  (it is a zwitterion). After irradiation, the stable species observed by ESR is a radical anion formed by deamination. Its formula is  $(\text{CH}_3)\cdot\text{CHCO}_2^-$ . It is trapped in the crystal lattice. A complete description of the radical reactions is based on the knowledge of the crystal structure and on the mechanisms observed in single crystals [23-25] or by spin trapping studies [26-27]. The mechanisms are similar in the two cases, and we can suppose in the first approach that in alanine powder, we have a large number of micro-crystals, so the reaction should not be different. But we must take into account that the specific area is very large and that the reaction at the surface and the influence of the neighbouring crystals for hydrogen bonds may modify the kinetics or the basic scheme.

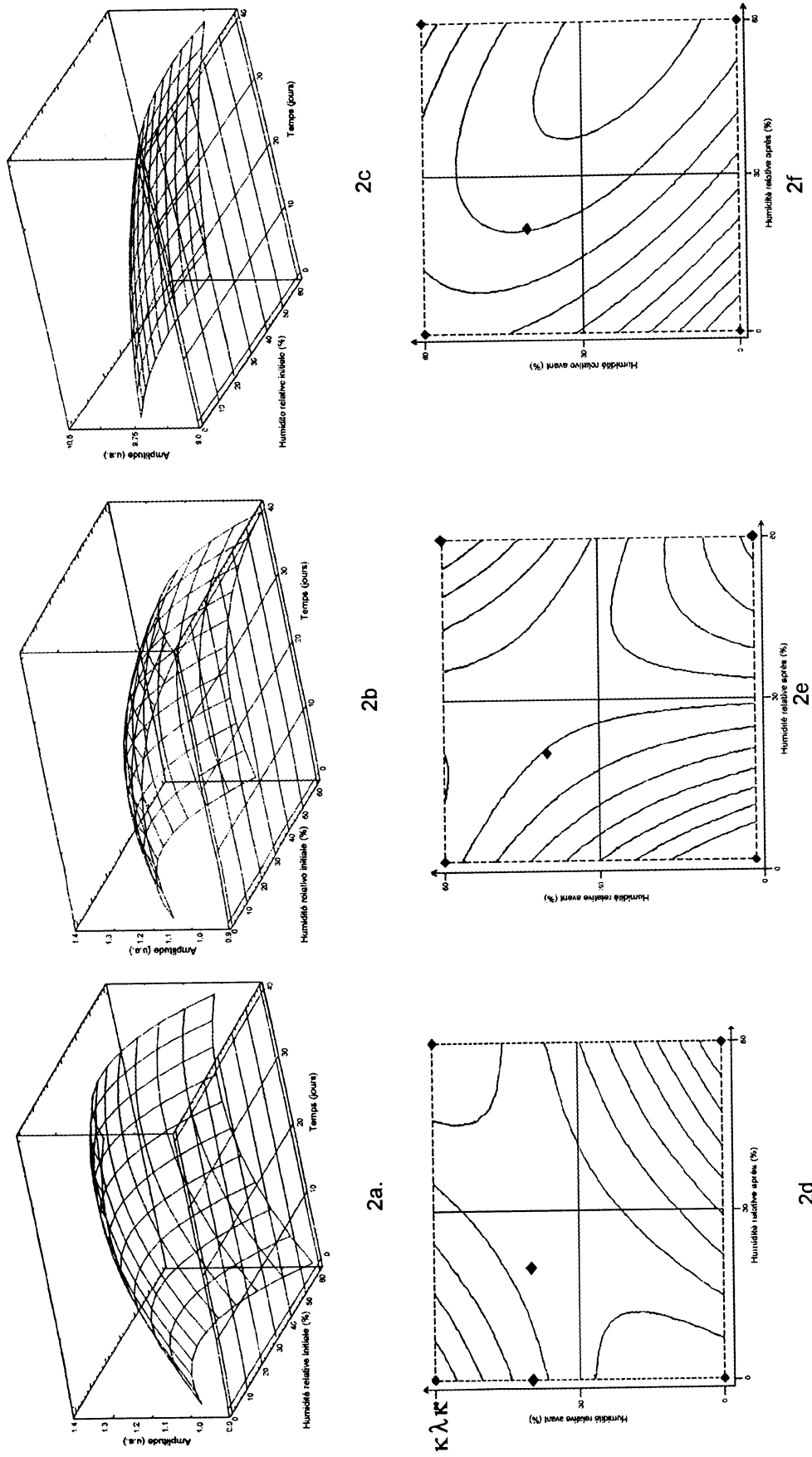


FIG. 2. Response surfaces and iso-signal functions for various humidity values and time after irradiation

- 2a : Response surface - powder, 1 kGy, 4 kGy/h, humidity for storage after irradiation 0%;
- 2b : Response surface - powder, 1 kGy, 4 kGy/h, humidity for storage after irradiation 60%
- 2c : Response surface - powder, 10 kGy, 4 kGy/h, humidity for storage after irradiation 60%
- 2d : Iso-signal curves - pellet, 10 kGy, 4 kGy/h, day 1
- 2e : Iso-signal curves - pellet, 1 kGy, 200 Gy/h, day 4
- 2f : Iso-signal curves - pellet, 100 Gy, 200 Gy/h, day 1

The first consistent description of the different steps of the reaction mechanism has been proposed by Minegishi et al [28] and more precisely developed by Shields et al [29]. The last two reactions of the mechanism should be intermolecular and/or intramolecular. They can exist at higher temperature than 77 K, but are poorly understood. Some authors [22, 23] describe the stable free radical in the single crystal as a planar species which can present three kinds of hydrogen bonds (two in the same plane and the third out of it) with neighbouring amino-protons. Those bonds are not equivalent for the radical formation process. It is well known that an electron is ejected by ionising radiation and is trapped by molecules to form the radical anion. The anion is trapped as a protonated radical, and protonation takes place across the hydrogen bridge. This proton located out of the plane is selectively transferred to the anion. The authors mention that three hydrogen exchange reactions may occur at room temperature.

We have worked with L- $\alpha$ -alanine powder (Merck) that is sieved without crushing to a granulometry between 150 and 180  $\mu\text{m}$ . The sample mass was about 100 mg and it was directly irradiated in a quartz "Suprasil" tube. The environmental conditions were ambient temperature and the relative humidity close to 0%. The powder was stored in a desiccator then transferred in the tube sealed with paraffin. A Philips X rays tube type MCN 321 was used. It delivers X photons with an average energy of 200 keV after filtration with 3 mm of aluminium. The experimental conditions of irradiations were 300 kV and 30 mA. The experiment was carried out, consisting of successive and cumulative irradiation steps, each irradiation being immediately followed by the ESR measurement. The irradiation times were first 5-minute steps followed by 12 steps of 10 minutes each. The dose rate was about 32.2 Gy/min, the total dose was around 4100 Gy. The small signal created by the quartz was not significant for the measurements of the alanine signal. After irradiation, the sample remains in the cavity of the spectrometer at room temperature and is measured at regular time steps, the measurements were performed during 8 days. The parameters used for the measurements were: frequency around 9.8 GHz, microwave power 1.3 mW, modulation amplitude 2.5 G, for a sweep width of 160 or 20 G.

In Fig. 3, we observe a curve close to a sigmoid. At the beginning, the measured signal of the free radical shows a weak slope and then increases linearly, and at the end a decrease of sensitivity. Figure 4 presents the evolution of the signal after irradiation. During the 8 days, it increases slowly but clearly more than 2%. The curve is characterised by a continuous increase with two different slopes and a stabilisation at the end. Many experimental points are affected by the thermal sensitivity of our ESR spectrometer diodes. These artefacts have been identified and these points (open circles) have been suppressed before using the data for modelisation.

For the analysis of these curves, assumptions can be made regarding the species which are involved and on the mechanism of reactions. The corresponding kinetic equations and curves can then be compared to experimental data. We consider that the observed increase in the signal after irradiation is probably due to several species not seen in our experiment and which would be transient species. We propose :



A is the alanine molecule, and B is a transient species. This species has not been seen by ESR measurements at room temperature in our experiments. R is the radical measured by ESR. It is often called secondary radical. The last reaction continues after irradiation.

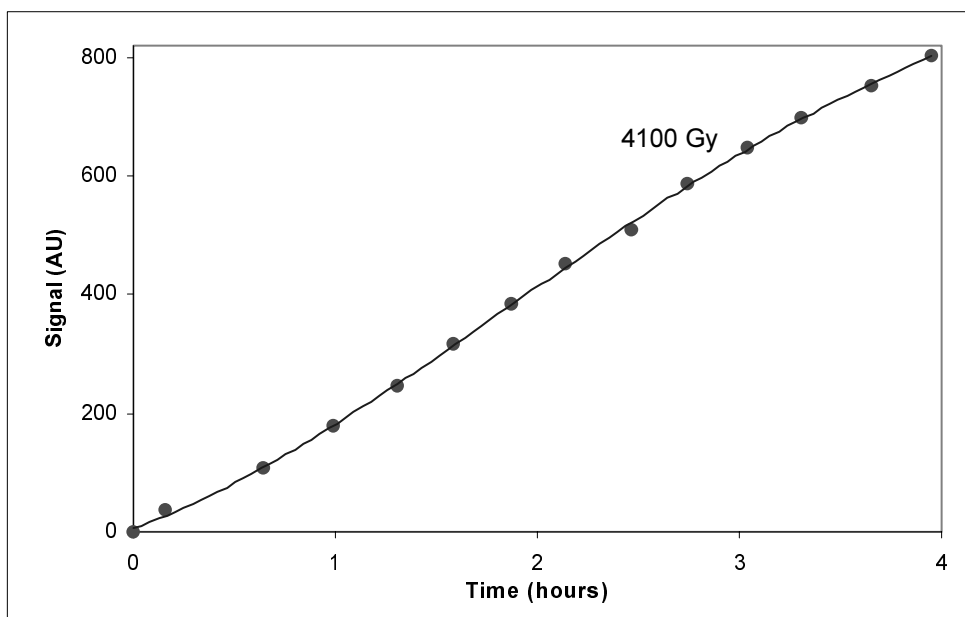


FIG. 3. ESR signal vs. elapsed time during successive irradiations

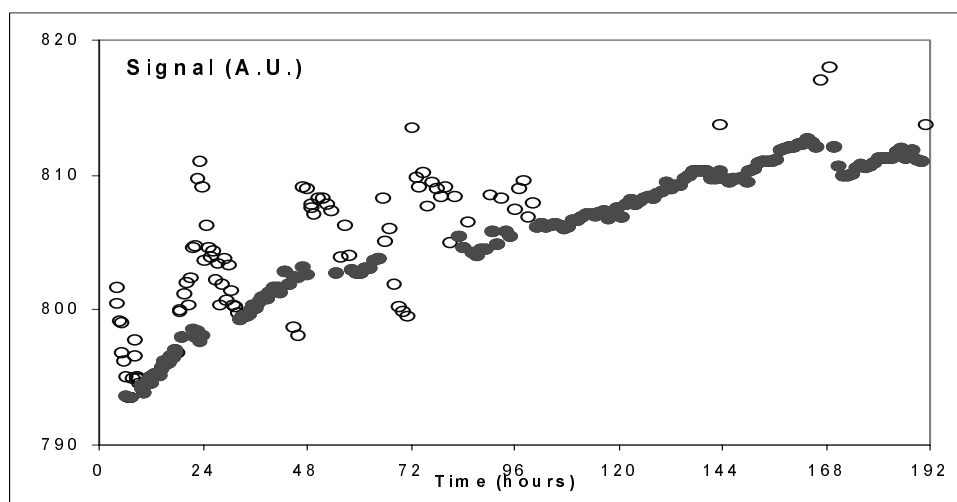


FIG. 4. Signal vs. time after irradiation (4100 Gy, 192 h). Solid points are data that were used for calculations

To analyse data in view of kinetics, regarding the two last reactions, a first order reaction is proposed as follows:

$$-d[B]/[B] = k \cdot dt \quad \text{which gives by integration : } \ln [(I_{\max}-I)/I_{\max}] = -k \cdot t$$



where,  $k$  is the apparent rate constant,  $I$  is the value of the ESR signal at time  $t$  of the secondary radical  $R$  corresponding to the disappearance of the transient species  $B$ , and  $I_{\max}$  is the maximum value of  $I$ .

We found a straight line and considered that it is true up to 120 hours. So, only these experimental points are used for the modelling. Regarding that conclusion, we have evaluated, after irradiation, the apparent rate constant by considering  $R_0$ , number of radicals existing at the end of irradiation, as initial condition. So we propose the equation :

$$R = DR_b \cdot (1 - e^{-kt}) + R_0$$

$DR_b$  represents the increase of the signal corresponding to the disappearance of the transient species,  $(DR_b + R_0)$  represents the limit value of the signal, and  $R_0$  may be considered as the initial value of the background immediately after irradiation. Figure 5 shows an accurate fitting and gives a value of the rate constant of about  $2 \times 10^{-2} \text{ h}^{-1}$ . These results suggest three conclusions. First, as it was shown [23–25] for a single crystal, a transient species also exists in powder. Because the signal increases after irradiation, at least one intermediate species and one reaction are likely. In the first approximation, the reaction mechanism can be considered identical to that in the single crystal. The transformation in a final stable radical takes place during and after irradiation. This mechanism is relatively slow and clearly observable at room temperature. Two other works suggest a mechanism of the formation of the final free radical. The study which has been performed by Sinclair and Hanna [30] at low temperature and on a single crystal of L-alanine suggests that the primary radiation effect is the ejection of an oxygen electron. This fact is characterised by an unpaired electron localised on the carboxyl group. This is confirmed by the work of Gottschall and Tolbert [31] on metal chelates of alanine. Sinclair and Hanna mentioned several steps for the radical reaction: the ejection of an electron leading to the formation of a positive ion and a negative ion. This negative ion is transformed into unstable free radical which gives, by direct deamination, the final stable radical. They suggested that the mechanism of decarboxylation of the positive ion is possible at low temperature creating an ethyl ammonium radical. Zagorski [32] suggested that the intermediate species, that he observed by absorption of light, is due to one or several precursors of the more stable ESR -detected species before deamination and decarboxylation. This intermediate species has been studied by pulsed radiolysis [33]. He measured the kinetics

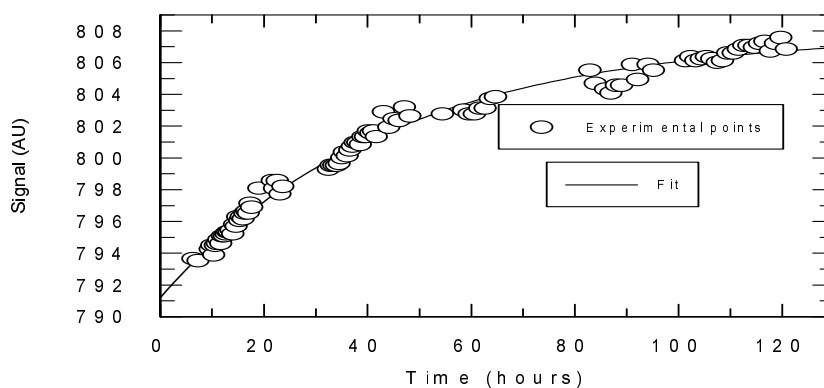


FIG. 5. Signal vs. time after irradiation (4100 Gy, 120 h): Comparison with fitted values

of formation and the decay of this intermediate species in a mono-crystal of L- $\alpha$ -alanine. Those approaches tend towards the same scheme: a simplification of the mechanism in two steps and at least one transient species. The first reaction having a very high rate constant, the correspondent species is only observable at low temperature. Pilbrow et al [34] put in evidence that, in irradiated alanine powder, two different free radicals might exist. But the irradiation conditions are not well defined. Makino et al [27] have studied irradiation of alanine in aqueous phases. They mentioned two parent radicals. The first radical ( $\text{CH}(\text{CH}_3)\text{COO}^-$ ) is formed by deamination reaction caused by the attack of aqueous electron, the second ( $\text{CH}_2\text{CH}(\text{NH}_3^+)\text{COO}^-$ ) by hydrogen abstraction reaction from the methyl group. For this last one, the observed spectrum is presented as the overlap of two forms: zwitterion and anion with a low interchange. We assume that kinetics are generally faster in aqueous than in solid phase and that the previously described reactions are probably similar in powder form with slower kinetics. Several researchers have studied the evolution of the ESR signal after irradiation [22, 35, 36]. They used in each case their own alanine pellets, thus making a meaningful comparison between these works difficult. These studies mention the influence of some parameters, such as temperature and humidity, without detailed information. From results presented in references [22, 36], we can suppose that apparent curves are the result of the competition between the increase due to the last reaction of formation of the observed radical and its disappearing by recombination or destruction in presence of water. In a previous work [37], we have shown, using an experimental design, that, depending on the experimental conditions used, results may be altogether very different and consistent.

## 5. CONCLUSION AND FUTURE DEVELOPMENTS

We have worked in three directions: the improvement of the signal (accuracy, repeatability, reproducibility), the understanding of the influence of the various external parameters by experimental design studies, and direct kinetics study and theoretical approach of the reaction mechanism.

The use of experimental designs is very satisfactory. Even though it is not easy to develop and to realise, the results give a general overview of the phenomenon with few experimentation. This method has permitted in several months to verify numerous results and to confirm the existence of some unexplained phenomenon. For example, a multiparametric approach has shown us favourable conditions for observing clearly an increase of the signal amplitude and then to prepare the kinetic study. The most marked evolution corresponds to the storage conditions after irradiation near 0% r.h. The presence of some other free radicals is a probability not negligible for this observation. We note an important effect of relative humidity before irradiation, which seems to condition the response since the first day of measurement, probably as the function of the equilibrium between the water and the alanine. We can suppose that the radiolysis of water and all the radicals created during the irradiation modify the chemical reactions significantly.

This work recommends first better specifications for the production and use of the alanine dosimeters, particularly for environmental conditions during all the life of the dosimeters till their ESR measurements. These conditions must be controllable and/or measurable for an accurate application of correction coefficients in the ESR normalisation of the signal. A second improved experimental design is in progress with some modifications, such as storage temperature as a controlled factor. Those hoped future results will permit us to give a better description of the kinetic evolution of the signal and its model.

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# CHARACTERIZATION OF FLUORIMETRIC, CALORIMETRIC, OSCILLOMETRIC AND RADIOCHROMIC DYE FILM DOSIMETERS UNDER PROCESSING CONDITIONS AT ELECTRON ACCELERATORS

A. KOVÁCS, M. BARANYAI, L. WOJNÁROVITS  
Institute for Isotope and Surface Chemistry, Chemical Research Center,  
Hungarian Academy of Sciences,  
Budapest, Hungary

## Abstract

Oscillometry was used for the evaluation of ethanol-monochlorobenzene (ECB) and aqueous alanine solutions. These solutions can be used in electron processing either in mylar pouches or glass ampoules for dose monitoring. The dose evaluation was carried out by using a new digital programmable oscillometric reader capable of measuring the dosimeter solutions stored in 1-ml and 2-ml glass ampoules. The temperature effect on the ECB dosimeters was tested in the temperature range of  $-30^{\circ}\text{C}$  to  $+50^{\circ}\text{C}$ . Fluorimetric analysis by measuring optically stimulated luminescence of solid and liquid phase organic and inorganic compounds for dose determination was used for characterization of Sunna dosimetric film containing LiF. A new thin polystyrene calorimeter was tested in the electron energy range of 1.5–10 MeV. GafChromic radiochromic dye films were also tested below 4 MeV and were found useful for dose determination for single pulse and scanned beam irradiations.

## 1. INTRODUCTION

Due to world wide growth of the radiation processing industry the role of quality assurance is a basic requirement. It requires the knowledge of combined external effects on the performance of reference and routine dosimeter systems used in gamma and electron irradiation facilities. At the same time the development and study of new dosimetry methods for use in radiation processing is also of basic significance. Special attention is required to develop new methods for use at medium and low energy electron accelerators as well as for bremsstrahlung (X rays) dosimetry.

The reliable application of dosimetry methods requires regular control with respect to accuracy and reproducibility compared to reference standard systems. Therefore the execution of intercomparison programmes aiming at comparison of the performance of routine systems to reference ones at different types of electron irradiation facilities is of basic significance.

These requirements constitute the objectives of the present Co-ordinated Research Project and our investigations aimed at the development of both existing and new dosimetry methods to be used for quality assurance of the radiation process. We have carried out research to improve the oscillometric analytical method to evaluate irradiated ethanol-monochlorobenzene and alanine dosimeter solutions. A thin polystyrene calorimeter was used for calibration and dose intercomparison studies especially at electron energies below 4 MeV. The applicability of the GafChromic film dosimeter and a new dosimetry system, the Sunna film have been tested for electron dosimetry purposes.

## 2. EXPERIMENTAL

### 2.1. Chemicals and dosimeters

The ethanol-monochlorobenzene dosimeter solution (ECB) was prepared and used as described elsewhere [1]. The naphthalene-2-carboxylic acid of reagent grade was dissolved in aqueous 0.1 M NaOH solution without further purification for OSL dosimetry.

GafChromic dosimeter films (DM-1260, MD-55 and NMD-55) produced by Gaf Chemicals Corporation [2] were tested and applied for 4 MeV electron irradiations using both single pulses and scanned beam. A new polymeric thin film developed by Sunna Systems Corporation [3] was studied in our laboratory for routine dosimetry in electron processing using both fluorimetric and spectrophotometric evaluation. The response of the films was determined both with gamma and electron radiation and at each dose point 5 film samples were irradiated and evaluated.

The humidity of the films was controlled using LiCl (12%),  $\text{MgCl}_2 \times 6 \text{H}_2\text{O}$  (34%),  $\text{Mg}(\text{NO}_3)_2 \times 6 \text{H}_2\text{O}$  (55%) and  $\text{KNO}_3$  (92%) solutions in polystyrene vials stored for 12 hours before irradiation. Water and polystyrene calorimeters designed and built at the High Dose Reference Laboratory of Riso National Laboratory were used for dose intercomparison studies. Measurement uncertainties, if not stated, are taken at 1 standard deviation.

### 2.2. Instrumentation

The *oscillometric* read-out method to evaluate the irradiated ECB solution was carried out with the OK-302/2 type oscillotitrator of Radelkis (Budapest, Hungary) and with the new, digital, programmable oscillometric reader developed by Sensolab Ltd. The *fluorimetric* measurements were performed partly with the Perkin-Elmer LS5 Luminescence Spectrometer to take the emission and excitation spectra of the samples studied and partly with the routine fluorimeter developed recently for the evaluation of the Sunna dosimeter films. The *spectrophotometric* measurements were performed with the Jasco V550 type UV-VIS spectrophotometer.

### 2.3. Irradiation facilities

The calibration and the irradiation of the dosimetry systems studied were carried out partly with the SLL-01 type  $^{60}\text{Co}$  gamma irradiation facility of the Institute of Isotopes Co. Ltd (Budapest, Hungary; nominal activity: 3 PBq) and with the gammacell type irradiation facility of the High Dose Reference Laboratory of Riso National Laboratory (Roskilde, Denmark; nominal activity: 8.1 TBq) in the temperature range of 10 to 55°C. Electron irradiations were performed with the 10 MeV linear accelerator (HRC-712 type, Haimson Research) of the High Dose Reference Laboratory of Riso National Laboratory (Roskilde, Denmark); with the 4 MeV linear accelerator (LPR-4 type, Tesla Vuvet, Czech Republic) of the Institute for Isotope and Surface Chemistry (Budapest, Hungary) and with the 2 MeV accelerator (GJ-2 type, Shanghai, China) of the Centre for Application of Isotopes and Radiation (Jakarta, Indonesia).

### 3. RESULTS AND DISCUSSION

#### 3.1. Development of oscillometric analysis

Oscillometry, i.e. the high-frequency method of chemical analysis to measure or follow changes in the composition of chemical systems, is used to evaluate irradiated ECB or aqueous alanine solutions. A new digital oscillometric reader with a sample holder for 2-ml dosimeter ampoules was designed and constructed recently [4]. According to our recent modifications the measuring frequency of the reader was changed to 85 MHz from 48 MHz and it is now capable of measuring absorbed doses in the range of 1–300 kGy extending its previous range of applicability above 100 kGy. The new reader consists of a built-in software to calculate the mathematical function of the calibration curve and thus to calculate the absorbed dose for the dosimeter samples irradiated in the dose range mentioned above.

#### 3.2. Temperature coefficient for ECB dosimetry

The effect of irradiation temperature on the oscillometric response of the ECB solution was investigated in the temperature range of  $-30^{\circ}\text{C}$  to  $+50^{\circ}\text{C}$  irradiating (in gamma field) ECB dosimeters to 1, 10 and 30 kGy nominal doses (see Table I).

TABLE I. IRRADIATION TEMPERATURE DEPENDENCE OF THE ECB SOLUTION USING OSCILLOMETRIC EVALUATION ( $1\sigma$ )

Irradiation temperature ( $^{\circ}\text{C}$ )	Absorbed dose* (kGy)	Absorbed dose* (kGy)	Absorbed dose* (kGy)
-30	$0.99 \pm 0.04$	$9.23 \pm 0.07$	$29.0 \pm 0.33$
0	$0.98 \pm 0.04$	$9.44 \pm 0.17$	$29.7 \pm 0.51$
25	$0.796 \pm 0.04$	$9.48 \pm 0.09$	$30.0 \pm 0.46$
50	$0.97 \pm 0.05$	$9.78 \pm 0.20$	$30.2 \pm 0.63$

\* The dose values are based on the calibration taken at  $25^{\circ}\text{C}$ .

No significant temperature effect was observed on the oscillometric response of the solutions as compared to the response at room temperature, i.e. the variation over the entire temperature range was not more than  $\pm 5\%$  ( $1\sigma$ ) and a temperature coefficient of  $0.015 \text{ kGy}/^{\circ}\text{C}$  was calculated at 30 kGy dose.

#### 3.3. Development of a thin polystyrene calorimeter

A new, thin polystyrene calorimeter was designed and built at the High Dose Reference Laboratory of Riso National Laboratory for dose measurements at lower electron energies, i.e. below 4 MeV [5]. This calorimeter has been tested in the 1.5–10 MeV electron energy range, since according to our depth-dose experiments the calorimeter was not totally absorbing even at 1.5 MeV energy. The main characteristics and the performance of this calorimeter was studied in detail in the frame of another Research Agreement (8530/CF) of this CRP, thus our measurements which have been carried out together with A. Miller, chief scientific investigator of that Research Agreement, are discussed in his report (elsewhere in this Annex).

### 3.4. Applicability of GafChromic dosimeter films at low electron energies

A new radiochromic film family consisting of a thin, radiation-active, microcrystalline dispersion coated on a polymer base has been developed during the past ten years with broad applications in radiation processing, food irradiation, clinical dosimetry, radiation and environmental protection. Different types of GafChromic films, i.e. DM-1260, MD-55 and NMD-55 have been tested for radiation processing applications mainly at 4 MeV electron irradiations. The dosimeter films were calibrated in  $^{60}\text{Co}$  gamma fields in two dose ranges, 30–200 Gy and 0.1–2 kGy, using different wavelengths (633 nm and 670 nm, respectively) for evaluation. These calibrations were then used to evaluate the irradiated GafChromic dosimeter films when applied at 4 MeV electron irradiations.

DM-1260 dosimeters were applied to measure absorbed dose in the dose range of 0.5–11 kGy using electron pulses of 2.6  $\mu\text{s}$  pulse length with 50-Hz repetition rate. The absorbed dose was also measured with ECB solution using the same irradiation conditions. (The ECB solution, calibrated in gamma field, was shown previously to be useful for electron dosimetry giving almost the same response as in gamma field). The difference between nominal dose (as calculated from ECB results) and measured dose (as determined from the GafChromic film readings at 400 nm) was within  $\pm 6\%$  ( $1\sigma$ ).

GafChromic films were also irradiated together with low-energy polystyrene calorimeters using the same irradiation geometry as that of the calorimeter. In this case scanned electron beam was applied (1-Hz scanning frequency, 25-cm scan width) and the nominal dose was measured with the calorimeters. The polystyrene phantom containing 5 pieces of GafChromic films was irradiated together with two polystyrene calorimeters, i.e. one before and one after the phantom. The average nominal dose as measured with the calorimeters was  $2.50 \pm 0.02$  kGy, while the GafChromic films measured  $2.45 \pm 0.04$  kGy.

These results indicate that GafChromic film dosimeters calibrated in gamma fields can be applied to measure absorbed doses in electron fields with a suitable accuracy ( $\pm 5\%$  at  $1\sigma$  level). It is necessary, however, to take into account the potential differences concerning the irradiation conditions during calibration and practical irradiation in order to carry out corrections if needed. In our case the calibration and electron irradiations were carried out at room temperature and the GafChromic films were packed into double layer sachets.

### 3.5. Dosimetry systems based on optically stimulated luminescence (OSL)

Fluorimetry is based on the measurement of fluorescent light, when a molecule — excited by UV or visible light — emits part of its energy in the form of light while getting back into ground state. The intensity of the fluorescent light — which is related to the concentration of the fluorescent compound — is measured with fluorimeters. This instrument consists of a source of light, a filter or monochromator, a holder for the sample and the detector (photomultiplier or photodiode).

Aromatic carboxylic acids have been shown earlier to be potential systems for dosimetry application due to the radiation induced formation of hydroxylated products as optically stimulated luminescent (OSL) agents. Detailed investigations have been carried out in this context with the aqueous solution of naphthalene-2-carboxylic acid and reproducible OSL response was found in the dose range of 0.1–100 kGy using a solute concentration of



minimum 0.01 M. Due to dose rate effects observed during characterization of the solution, however, its potential use as a dosimeter is limited, unless suitable additives to eliminate this effect can be applied.

The characterization of a new potential dosimetry material, the Sunna film [3, 6] has been carried out with respect to its use in electron irradiated fields. This film contains an inorganic OSL fluor (LiF) dispersed in a polymer matrix. The evaluation of the irradiated films can be carried out with OSL or with spectrophotometric analysis. When using OSL measurements a new, table-top routine fluorimeter was used for this purpose, which excites the dosimetric material at 450 nm and measures the emitted light at 670 nm. The reader developed by Sensolab Ltd is equipped with a convenient software to store data to calculate calibration function by fitting the response curve with a polynomial or linear regression function and to deliver measured and calculated data to a PC. The spectrophotometric analysis is carried out at 240 nm using relatively wide slitwidth (10 nm) and measuring the absorbance of the irradiated film against the unirradiated one.

The response of the Sunna film was found different in the case of gamma and electron irradiation both for OSL and spectrophotometric analyses. Figure 1 shows the response of the Sunna film with OSL evaluation, while the response with spectrophotometric evaluation is shown in Fig. 2 [6, 7]. Therefore the calibration of the film has to be carried out separately unlike e.g. in the case of the GafChromic films as discussed earlier. The intrabatch non-uniformity ( $1\sigma$ ) of the film was found less than 2% [3].

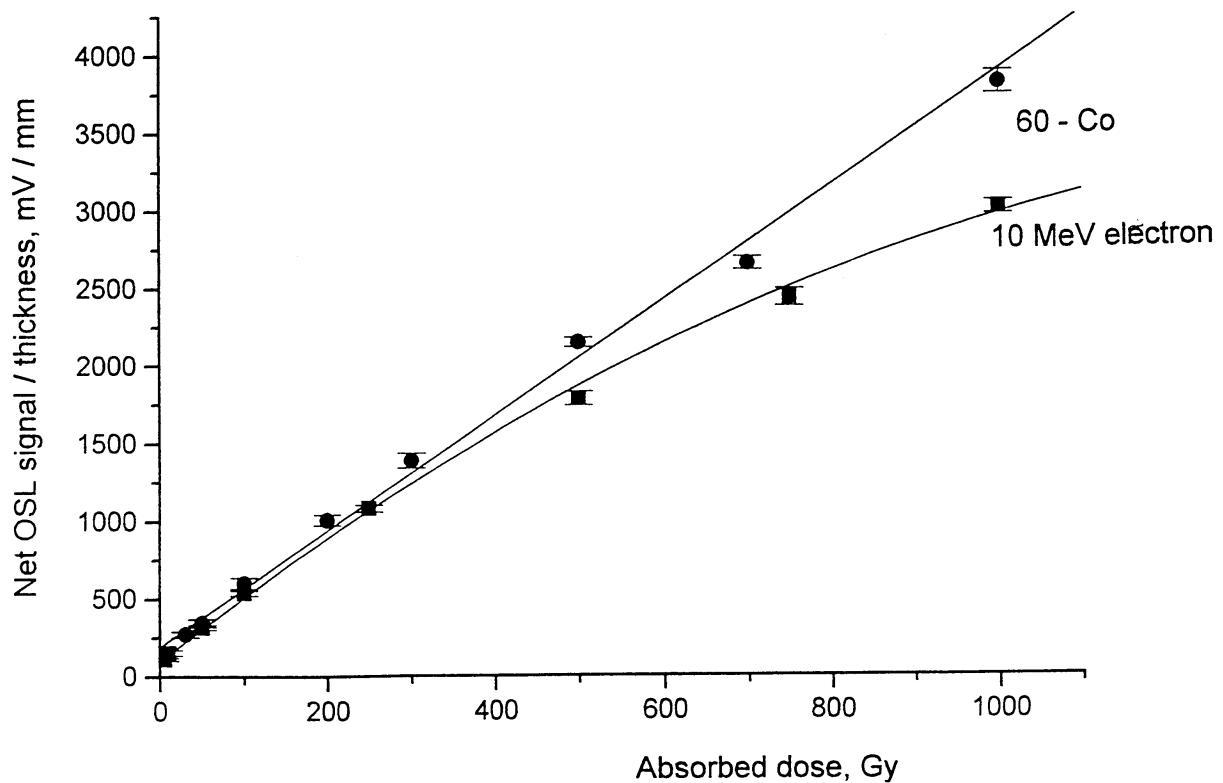


FIG. 1. Electron and gamma dose response of the Sunna dosimeter film using OSL evaluation (Uncertainty:  $1\sigma$ ).

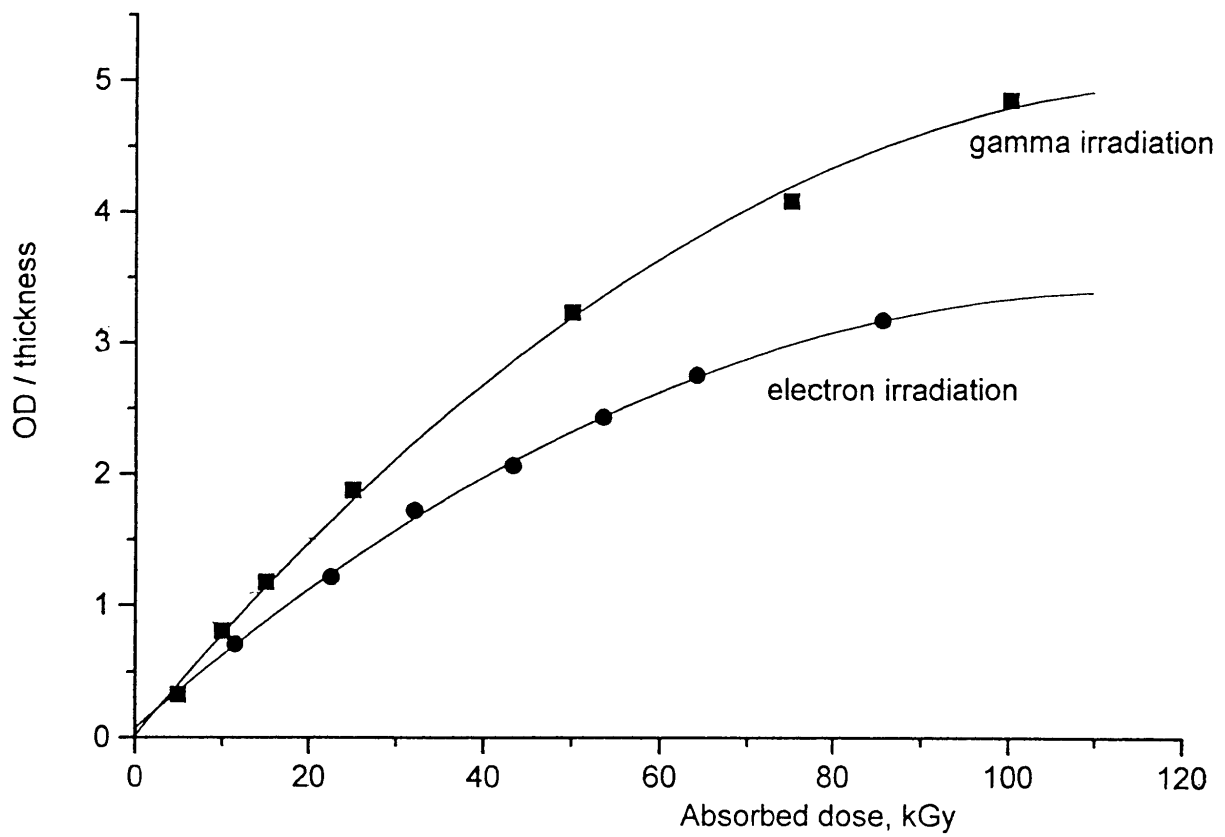


FIG. 2. Electron and gamma dose response of the Sunna film using spectrophotometric evaluation, (Uncertainty:  $1\sigma$ ).

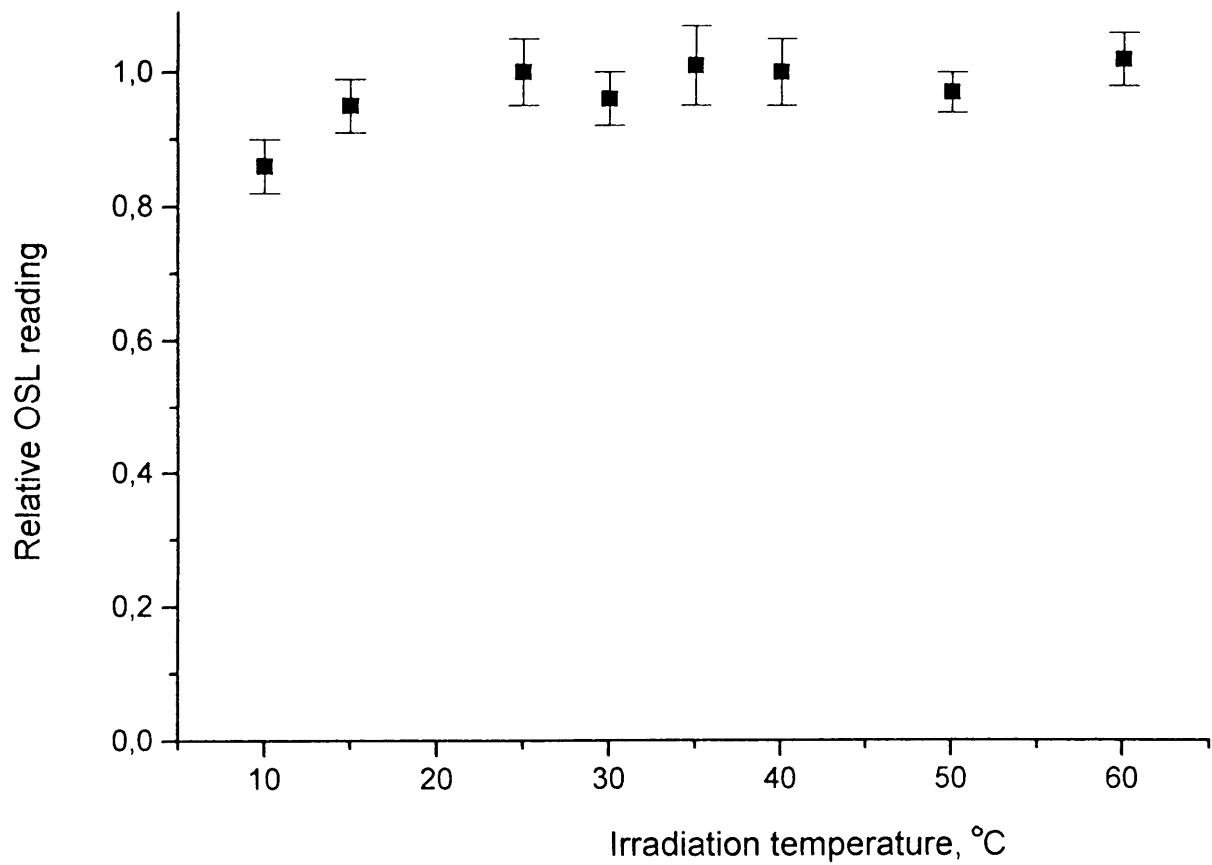


FIG. 3. Effect of irradiation temperature on the dose response of the Sunna dosimeter film (All data are normalized to the OSL value measured at  $25^{\circ}\text{C}$ ; Uncertainty:  $1\sigma$ ).

The effect of relative humidity (in the range of 12–92%) at 15, 25, 35 and 55°C temperatures was also studied irradiating the films to 5 kGy. As seen in Table II the net

TABLE II. EFFECT OF RELATIVE HUMIDITY ON THE  $\Delta$ OSL/THICKNESS VALUES OF THE SUNNA FILM

Temperature, °C	12% r. h.	34% r. h.	55% r. h.	92% r. h.
15	2424	2521	2525	2522
25	3146	3083	3050	3028
35	2568	2297	2380	2303
55	1535	1546	1488	1576

### 3.5.1. Characteristics of the OSL analysis

The Sunna film has been found suitable for dose measurements in the range of 10 Gy–100 kGy using OSL analysis. However, detailed investigations on its response have shown significant irradiation temperature effect at doses above 5 kGy in the temperature range of 10–60°C, when using OSL analysis. Below this dose level, however, insignificant temperature effect on the OSL signal of the Sunna film was found between 20°C and 60°C, while a slight decrease was observed below 20°C (see Fig. 3).

$\Delta$ OSL/thickness values changed only slightly with the increasing humidity at different temperatures.

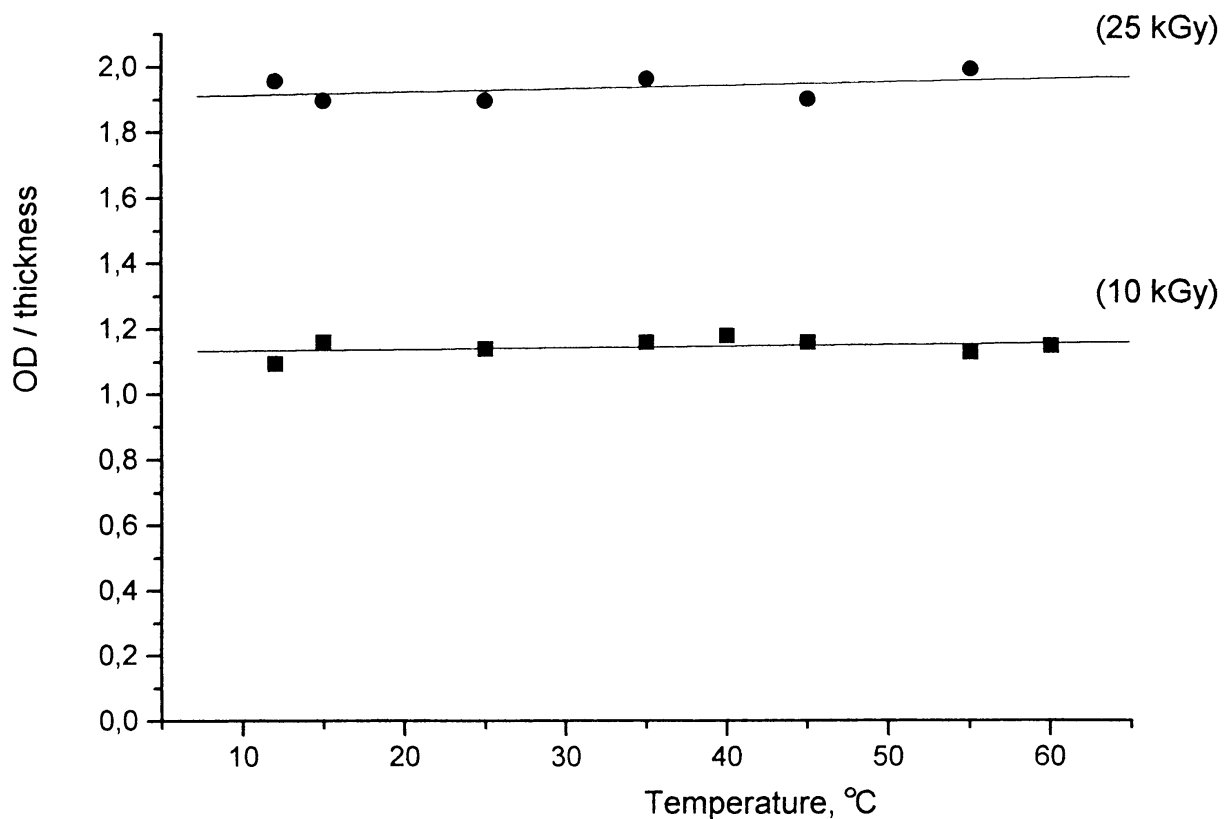


FIG. 4. Irradiation temperature effect on the absorbance of the irradiated Sunna film.

### 3.5.2. Characteristics of the spectrophotometric analysis

The effect of irradiation temperature was studied when spectrophotometric evaluation was carried out by measuring the absorbance of the radiation induced colour centers at 240 nm in the case of films irradiated to doses higher than 5 kGy. No significant variation of response was observed in the temperature range studied at 10 kGy and 25 kGy dose levels (see Fig. 4).

The effect of relative humidity was also investigated on the spectrophotometric response, ( $\Delta$  absorbance/thickness) of the Sunna film at different temperatures as shown in Table III. and no significant effect was observed except at 92% relative humidity.

TABLE. III. EFFECT OF RELATIVE HUMIDITY ON THE SPECTROPHOTOMETRIC RESPONSE OF THE SUNNA FILM

Temperature, °C	12% r. h.	34% r. h.	55% r. h.	92% r. h.
15	0.788	0.784	0.800	0.857
25	0.833	0.826	0.867	1.011
35	0.742	0.702	0.740	0.901
55	0.828	0.815	0.830	0.982

## 4. CONCLUSIONS

We have carried out investigations to develop and to test both existing and new dosimetry systems to be used in radiation process control. These investigations were in accordance with the main goals of this CRP, i.e. to propose, develop and test transfer and routine dosimeters in use especially at electron accelerators of energies below 4 MeV.

GafChromic films were found to be suitable for electron dose determination based on calibration carried out in gamma radiation fields.

The introduction of the polymer based LiF film (the Sunna film) has been found applicable for dose determination in a wide dose range. Fluorimetry is suggested to be applied for dose measurement from 10 Gy up to about 5 kGy [3, 6], while higher doses up to 100 kGy can be determined by spectrophotometry [6]. The characterization of this new system resulted in the knowledge of the influence of various external parameters leading to the proposition of using two different evaluation techniques. Since critical environmental conditions (such as, humidity and under certain conditions irradiation temperature) showed no effect on the response of the dosimeter film [6] and its thickness is 0.45 mm, it seems to be a suitable choice for dose monitoring in electron radiation processing even at low (~1 MeV) electron energies.

## ACKNOWLEDGEMENTS

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# USE OF SPECTROPHOTOMETRIC READOUT METHOD FOR FREE RADICAL DOSIMETRY IN RADIATION PROCESSING INCLUDING LOW ENERGY ELECTRONS AND BREMSSTRAHLUNG

B.L. GUPTA

Chemical Dosimetry Group,  
Bhabha Atomic Research Centre,  
Trombay, Mumbai, India

## Abstract

Our laboratory maintains standards for high doses in India. The glutamine powder dosimeter (spectrophotometric readout) is used for this purpose. Present studies show that 20 mg of unirradiated/irradiated glutamine dissolved in freshly prepared 10 ml of aerated aqueous acidic FX solution containing  $2 \times 10^{-3} \text{ mol dm}^{-3}$  ferrous ammonium sulphate and  $10^{-4} \text{ mol dm}^{-3}$  xylenol orange in  $0.033 \text{ mol dm}^{-3}$  sulphuric acid is suitable for the dosimetry in the dose range of 0.1–100 kGy. Normally no corrections are required for the post-irradiation fading of the irradiated glutamine. The response of glutamine dosimeter is independent of irradiation temperature in the range of about 23–30°C and at other temperatures, a correction is necessary. The dose intercomparison results for photon, electron and bremsstrahlung radiations show that glutamine can be used as a reference standard dosimeter. The use of flat polyethylene bags containing glutamine powder has proved very successful for electron dosimetry of wide energies. Several other amino acids like alanine, valine and threonine can also be used to cover wide range of doses using spectrophotometric readout method.

## 1. INTRODUCTION

Bhabha Atomic Research Centre has been accredited as a National Calibration Laboratory in the field of radiological measurements. The absorbed dose in the dose range of 0.1 to 20 Gy is calibrated with a graphite ionization chamber using IAEA's TRS 277 protocol. The absorbed doses in radiation processing dose range are calibrated against Fricke dosimeter. Table I gives radiation processing facilities available in India and the purpose for which they are used. Alanine, glutamine, threonine and several other amino acid free radical dosimeters in powder form with a spectrophotometric readout are used for measurement of dose in radiation processing dose range and for transfer dosimetry. The sensitive FBX dosimeter which was developed in our laboratory is also used for dose calibrations in the dose range of 0.01 to 50 Gy. Atomic Energy Regulatory Board (AERB) which is the licensing authority for food irradiation plants has assigned the dose assurance responsibility in food irradiation plants to our laboratory [1]. The dose assurance in radiotherapy is carried out by the Secondary Standards Laboratory in our Division (Radiation Standards and Instrumentation Division) at BARC. In food irradiation, the overall uncertainty of delivered dose in routine food irradiation shall not exceed 10% at  $1\sigma$  level. An agreement in the range of 6% at 95% confidence level in dose intercomparisons carried out under controlled condition of irradiation is considered desirable.

## 2. GLUTAMINE DOSIMETER

For the dosimetry in radiation processing dose range, glutamine dosimeter using spectrophotometric readout has proved very useful. In this method, 20 mg of irradiated glutamine was being dissolved in 10 ml of an aerated aqueous solution containing  $2 \times 10^{-4} \text{ mol dm}^{-3}$  ferrous ammonium sulphate and  $10^{-4} \text{ mol dm}^{-3}$  xylenol orange in  $0.033 \text{ mol dm}^{-3}$  sulphuric acid (FX solution). Ferrous ions are oxidized by the peroxy radicals produced from

TABLE I. RADIATION PROCESSING PLANTS IN INDIA

Name	Max. capacity (kCi)	Application
ISOMED, Mumbai	1000- <sup>60</sup> Co	Medical product sterilization
RASHMI, Bangalore	300- <sup>60</sup> Co	Medical product sterilization
SARC, Delhi	500- <sup>60</sup> Co	Medical product sterilization & food irradiation
RAVI, Jodhpur	300- <sup>60</sup> Co	Medical product sterilization, food irradiation and others
FTD, BARC	100- <sup>60</sup> Co	Food irradiation
RUBBER BOARD, Kottayam	300- <sup>60</sup> Co	Rubber latex vulcanisation
SHRI, Vadodara	500- <sup>60</sup> Co	Sludge hygienisation
Isotope Division, BARC	2 MeV electrons	General purpose
Isotope Division, BARC (PANBIT)	100- <sup>60</sup> Co	General purpose
Shree Chitra Institute, Trivandrum (PANBIT)	100- <sup>60</sup> Co	Medical product sterilization
Western India Plywood, Baliapatnam (PANBIT)	100- <sup>60</sup> Co	Wood polymer
NRL, BARC, Srinagar (PANBIT)	100- <sup>60</sup> Co	Medical product sterilization
BRIT, New Mumbai	1000- <sup>60</sup> Co	Spices irradiation
Poton irradiator, Nashik	100- <sup>60</sup> Co	Onions & potatoes Irradiation
BRIT, New Mumbai	500 keV electrons	Surface curing & others
Mangalore University, Mangalore	8 MeV electrons & Bremsstrahlung	Multipurpose

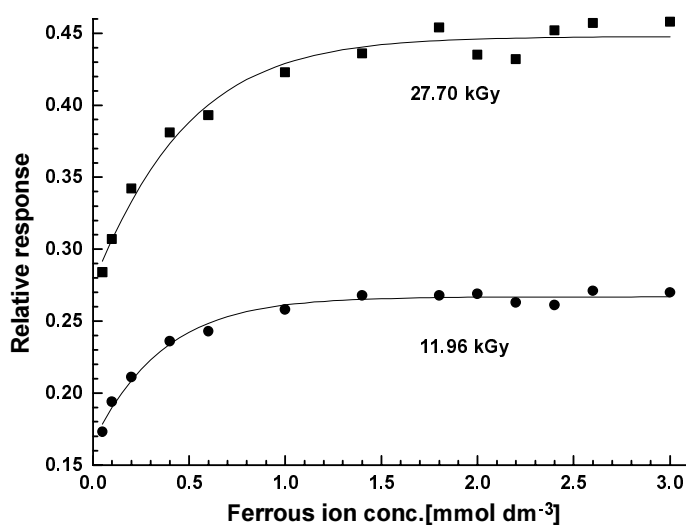


FIG.1. Effect of ferrous ion concentration in FX solution on dose estimation by glutamine dosimeter.



irradiated glutamine. Xylenol orange forms a complex with ferric ions as well as controls the chain length of ferrous ion oxidation. Effect of ferrous ion concentration in this method was investigated. As shown in Fig.1,  $2 \times 10^{-3}$  mol dm<sup>-3</sup> ferrous ammonium sulphate concentration is required for optimal oxidation of ferrous ions. Now this concentration is used for FX solution preparation for glutamine dosimeter.

### 3. EFFECT OF FX STORAGE TIME ON DOSE EVALUATION BY GLUTAMINE DOSIMETER

The FX solution used for glutamine dosimeter is allowed to stabilize for about an hour before use. After dissolution of the irradiated/unirradiated glutamine in FX solution, the solution is kept for about 30 minutes for the reaction to complete. The absorbance of both the irradiated and unirradiated glutamine solutions is measured at 549 nm against the FX solution. The ferrous ions undergo thermal oxidation during this period. Earlier, we have used  $2 \times 10^{-4}$  mol dm<sup>-3</sup> of ferrous ammonium sulphate in FX solution and thermal oxidation of the two solutions did not introduce any error. The increase of ferrous ion concentration in Section 2 in FX solution increases the rate of thermal oxidation of ferrous ions. Therefore, the effect of storage time of FX solution containing increased concentration of ferrous ions, after its preparation on dose evaluation by glutamine was studied. Since this method of dose estimation is based on the FBX dosimeter developed by us and the storage of FBX solution at about 10°C considerably reduces the rate of thermal oxidation of ferrous ions, studies were done using the storage temperatures of 26°C (room temperature) and 10°C. FX solution stored at these two temperatures for different periods varying from 1 to 29 h were used to estimate the dose from glutamine dosimeters irradiated to three different doses. The same irradiated powder was used for the 8 dose estimations 4 for each temperature. The results in Table II show that the FX solution once prepared can be stored for about 24 hours. However, there is some uncertainty for very high doses. For good accuracy it is advisable to prepare a fresh FX solution and use it the same day for the dose estimation.

TABLE II. EFFECT OF FX STORAGE TIME ON DOSE EVALUATION BY GLUTAMINE DOSIMETER

Temp. (°C)	Dose (kGy)	Dose (kGy) after storage time of			
		1 h	6 h	24 h	29 h
26.0	8.85	8.83	8.83	8.85	9.06
	31.00	30.95	30.95	31.10	31.55
	68.50	68.50	71.10	71.10	73.00
10.0	8.85	8.83	8.95	8.83	8.71
	31.00	30.95	30.64	30.79	30.95
	68.50	68.50	70.20	70.20	69.00

### 4. EFFECT OF IRRADIATION TEMPERATURE ON DOSE ESTIMATION BY GLUTAMINE DOSIMETER

Glutamine powder was irradiated to different doses at different temperatures in a gamma chamber. The irradiated glutamine dosimeters were evaluated using the spectrophotometric readout method and the above composition of FX solution was used for

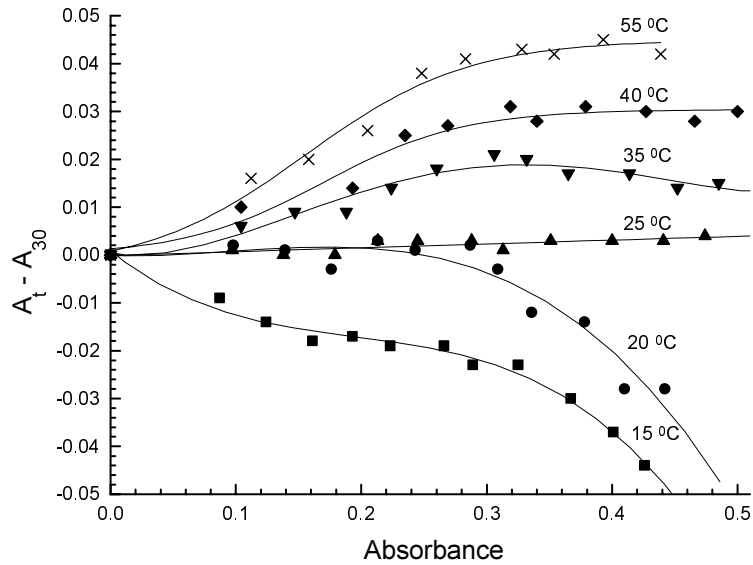


FIG. 2. Change in absorbance on irradiation of glutamine dosimeter at different temperatures.

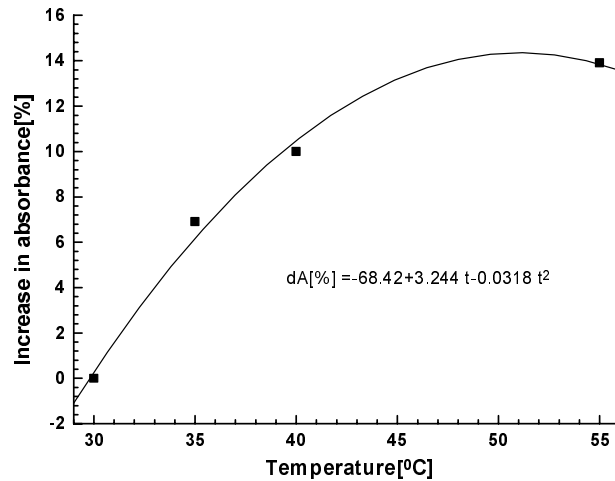


FIG. 3. Percentage increase in absorbance on irradiation of glutamine dosimeter at different temperatures.

this purpose. Figure 2 shows the difference in absorbance of glutamine dosimeter between  $t^{\circ}\text{C}$  and  $30^{\circ}\text{C}$  against the absorbance at  $t^{\circ}\text{C}$ . These results show that the effect of temperature during irradiation of glutamine dosimeter is minimum around  $30^{\circ}\text{C}$ .

The difference in absorbance for irradiations at  $30^{\circ}\text{C}$  and higher temperatures increases up to an absorbance value of about 0.3 which corresponds to 15 kGy and as seen from the figure, at any irradiation temperature, the increase in absorbance is almost linear with the absorbance (up to a value of 0.3) at the irradiation temperature. This increase ( $dA$ ) at different temperatures is given by the relation:  $dA[\%] = -68.42 + 3.24t - 0.031t^2$  where  $t$  is the irradiation temperature in  $^{\circ}\text{C}$  (Fig. 3). For absorbance values above 0.3, the increase in absorbance

remains almost constant. The difference in absorbance for irradiations at 30°C and lower temperatures up to about 20°C is almost zero up to 15 kGy. For both 15 and 20°C, the difference above 15 kGy is about 20% of the absorbance value above 0.3. When the irradiation temperature for doses below 15 kGy is above 30°C, the absorbance at irradiation temperature  $t^{\circ}\text{C}$  above 30°C is corrected and the dose is read from the calibration curve between 20–30°C. The plot of absorbance versus dose is non-linear and the plot of inverse of absorbance versus inverse of dose is almost linear (Fig. 4). It is advisable to use a polynomial fit of dose versus absorbance (Fig. 5) for more accurate results.

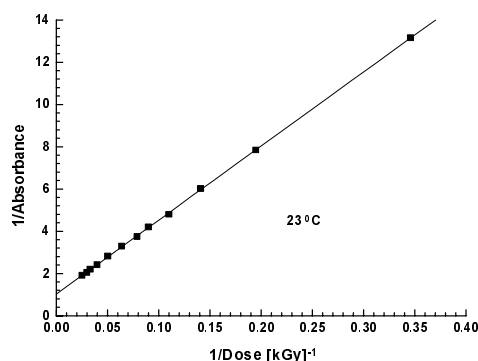


FIG. 4. Plot of inverse of absorbance versus inverse of dose for glutamine dosimeter.

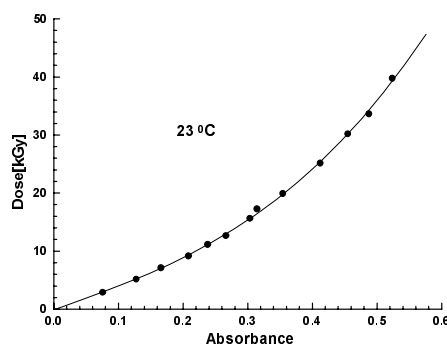


FIG. 5. Plot of absorbance versus dose for glutamine dosimeter.

## 5. POST-IRRADIATION FADING IN GLUTAMINE DOSIMETER

In India, the summer temperatures rise to about 50°C in some places. Therefore, glutamine dosimeters were irradiated to doses of 4.8, 19.6 and 57.9 kGy and stored at two temperatures of 25°C and 50°C. The post-irradiation stability of glutamine dosimeter was found to depend on dose and temperature of storage. At 25°C, the fading is negligible up to 4 months for doses of 4.8, and 19.6 kGy and it slowly increases to about 3.6% during this period for 57.9 kGy. At 50°C, the fading is negligible for 5 kGy up to 4 months, however, for 19.6 kGy, the fading is negligible up to 1 month and beyond that period, in about 4 months it increases to about 5%. For 57.9 kGy, the fading is less than 1% in about a month and it increases to about 9% in about 4 month time.

## 6. INTERCOMPARISON OF FRICKE DOSIMETER AND IONIZATION CHAMBER

In our Division, the Gamma Chamber is calibrated against the Fricke dosimeter. The teletherapy level dosimeters are calibrated against the graphite ionization chamber and the FBX dosimeter. Therefore, an intercomparison of the these methods was carried out. Dose measurements were done with a  $^{60}\text{Co}$  teletherapy machine in a water phantom at a depth of 5 cm. The dose-rate measured by both ionization chamber and FBX dosimeter was 31.3 Gy/h showing an excellent agreement. Table III gives the results of intercomparison between Fricke dosimeter and ionization chamber, again showing very good agreement [2].

Since the glutamine dosimeter is used for dose assurance programme in food irradiation plants in India, a detailed study was done about the uncertainties involved in dose

measurement by glutamine dosimeter. Table IV gives the uncertainties in dose measurement by glutamine dosimeter. A bottle of glutamine contains about 100 g powder which is considered as a batch for calibration purposes. A bottle lasts for a long time. There are no variations in the batch since the powder is quite homogeneous.

TABLE III. INTERCOMPARISON OF FRICKE DOSIMETER AND IONIZATION CHAMBER

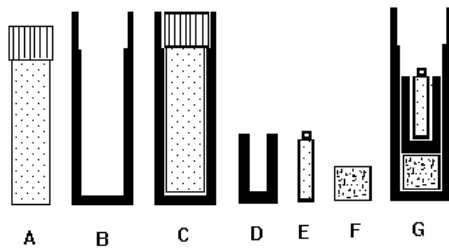
Expt No.	Dose rate(Gy/h)		Fricke/Ion. chamber
	Fricke	Ionization Chamber	
1	31.08	30.96	1.004
2	30.92	30.96	0.999
3	30.78	30.89	0.996
4	30.73	30.89	0.995

TABLE IV. UNCERTAINTIES IN GLUTAMINE DOSIMETER CALIBRATION AT  $1\sigma$  (%)

Component	$1\sigma$ (%)
Dose-rate by Fricke dosimeter	0.7
Difference in Fricke and glutamine size	0.6
Calibration formula for glutamine	1.1
Post-irradiation stability	0.3
Temperature correction	0.5
Total	1.6

## 7. IRRADIATION GEOMETRY FOR CALIBRATION

Because of the variation in beam intensity and electron scatter conditions, a proper geometry is required for accurate dosimetry. Figure 6 gives the holders designed and used in our laboratory for calibration of free radical dosimeters using the Fricke dosimeter as reference standard. A build-up of about 5 mm thickness of unit density material is necessary to avoid the contribution of electrons generated from high atomic number materials in a gamma chamber. Figure 7 shows the arrangements when either one or three dosimeters are used for the calibration. In a process load, the product may not be uniformly distributed. Therefore, for a reliable intercomparison, it is better to use a compact holder for placing the dosimeters. The dosimeters should be vertical and perpendicular to the beam direction to avoid dose variations over the dosimeter volume. Figure 8 shows an arrangement to achieve this goal for dosimetry in a process load. It is advisable to place dosimeters on a sheet so that the positions of the dosimeters can be properly arranged.



- A - Polypropylene tube for Fricke dosimeter
- B - Perspex build-up cap for Fricke dosimeter
- C - Polypropylene tube with perspex build-up
- D - Perspex build-up for glutamine dosimeter
- E - Polystyrene tube for glutamine dosimeter
- F - 13 mm dia and 13 mm height perspex spacer
- G - Glutamine dosimeter with perspex build-up

FIG. 6. Different holders for calibration of free radical dosimeters.

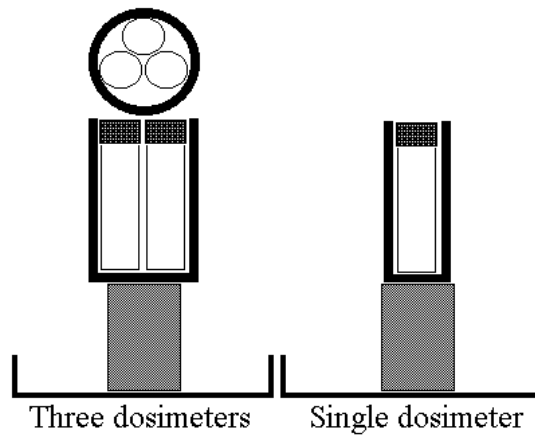


FIG. 7. Arrangement for calibration of free radical dosimeters.

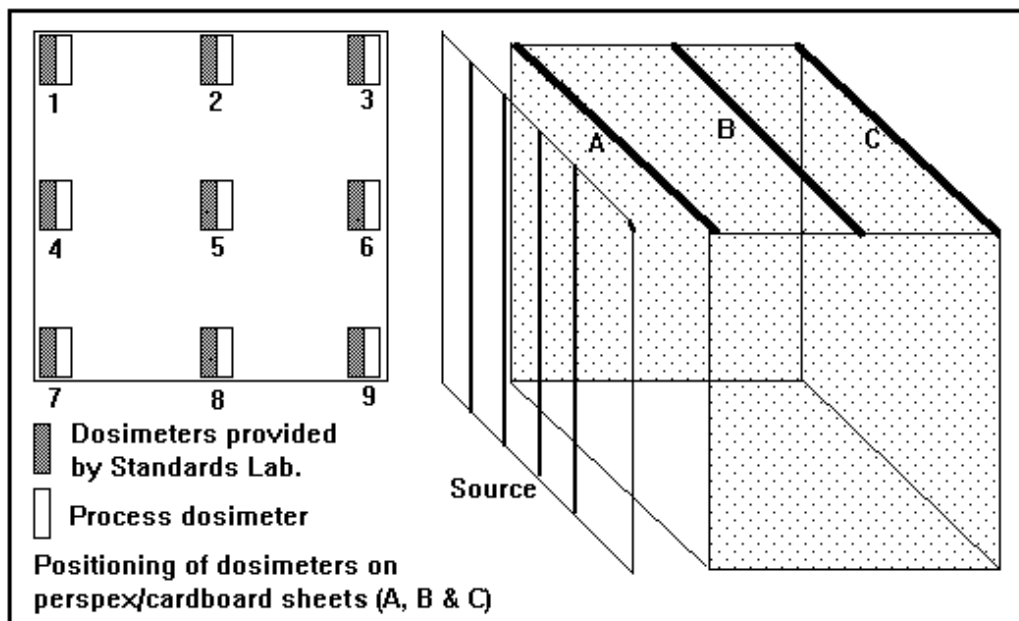


FIG. 8. Arrangement for dose variation study in a process load.

## 8. DOSE INTERCOMPARISON BETWEEN BARC AND NPL(UK)

Nine dichromate dosimeters (3 dosimeters for each dose) were irradiated in a gamma chamber at BARC. The BARC/NPL dose ratio was found to be  $0.983 \pm 0.006$  which shows a good agreement (see Table V). Nine glutamine dosimeters (3 for each dose) were irradiated at NPL in a gamma chamber at 25°C and they were read at BARC using the spectrophotometric readout method. Table VI gives the results of this dose intercomparison which shows an excellent agreement. The average ratio of BARC/NPL =  $0.997 \pm 0.010$  [3].

TABLE V. DOSE INTERCOMPARISON BETWEEN NPL AND BARC USING DICHROMATE DOSIMETERS

Dose(kGy) BARC NPL		BARC/NPL
13.6	13.91	0.978
23.2	23.54	0.985
36.8	37.32	0.986

TABLE VI. DOSE INTERCOMPARISON BETWEEN BARC AND NPL USING GLUTAMINE DOSIMETERS

Dose(kGy) BARC NPL		BARC/NPL
6.47	6.49	0.997
17.2	17.2	0.998
37.9	38.1	0.994

## 9. DOSE INTERCOMPARISON BETWEEN BARC AND IAEA

Three alanine (EPR) dosimeters received from IAEA were irradiated in a gamma chamber in two separate experiments in 1996 and 1997. Table VII gives the experimental details for the irradiation at BARC and Table VIII gives the results of this intercomparison. There is a very good agreement between the two values. Nine glutamine dosimeters (3 for each dose) were irradiated to three doses in a Gamma cell at the IAEA laboratory. The average of the ratios of doses measured by BARC and the doses given by IAEA is  $0.989 \pm 0.015$  (Table IX).

TABLE VII. EXPERIMENTAL DETAILS FOR IRRADIATION OF ALANINE AT BARC

Uncertainty ( $1\sigma$ ) in the dose value	Field uniformity	= 0.3%
Type A = 0.69%	Traceable to	NPL(UK)
Type B = 0.25%	BARC/NPL dose ratio	= $0.983 \pm 0.006$
Total = 0.73%		

TABLE VIII. DOSE INTERCOMPARISON BETWEEN BARC AND IAEA USING ALANINE (EPR) DOSIMETERS

	1996	1997
BARC nominal dose(kGy)	24.8	15.32
IAEA estimated dose(kGy)	25.08	15.26
Relative error	-1.11	0.39

TABLE IX. DOSE INTERCOMPARISON BETWEEN BARC AND IAEA USING GLUTAMINE DOSIMETERS

IAEA (kGy)	dose	BARC (kGy)	dose	BARC/IAEA
5.00		5.07		1.014
10.02		9.80		0.978
30.02		29.5		0.983

## 10. ELECTRON BEAM IRRADIATIONS

Extensive work was done by us in collaboration with Isotope Division of BARC on the dosimetry of 2 MeV industrial electron accelerator using glutamine dosimeter. The accelerator has a scanned beam (90 cm × 7 cm). The glutamine powder (5.2 mg/cm<sup>2</sup>) is filled in polyethylene bags (polyethylene wall thickness = 5.9 mg/cm<sup>2</sup>) and these bags are placed at different depths using cellulose triacetate film spacers in a wooden phantom. The density of glutamine is 1.54 gm/cm<sup>3</sup>. The bags are pressed by a cellotape of 2.5 mg/cm<sup>2</sup> thickness. After the irradiation, each bag is measured separately using the spectrophotometric readout method. The depth doses were measured at the centre of the beam and at 25 cm from the centre. In one experiment, the conveyor was stationary while in the second experiment, the conveyor was moving. Figure 9 gives the results of depth dose measurements. The maximum dose was found at a depth of 240 mg/cm<sup>2</sup>. Similar results were obtained with a moving conveyor.

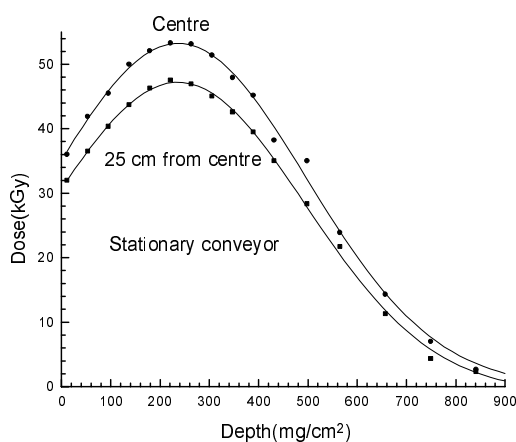


FIG. 9. Depth doses from a 2 MeV electron beam using glutamine dosimeter.

## 11. DOSE INTERCOMPARISON WITH RISO NATIONAL LABORATORY

Since glutamine filled in thin flat polyethylene bags is quite suitable for electron beam dosimetry, a dose intercomparison was carried out with Riso National Laboratory. In the first experiment done in March 1998, glutamine dosimeters (3 for each dose) were irradiated with 10 MeV electron beam. The results of this intercomparison are given in Table X showing a good agreement. The experiment was repeated in September 1998 both for <sup>60</sup>Co and 10 MeV electron beam. Alanine(EPR) dosimeters were used by RISO for dosimetry of gamma and electron beams. The results for both as given in Tables XI and XII show good agreement. Thus the gamma calibration can be used for electron beam without any correction. However, a care is required for temperature variations during the electron beam irradiations since these variations may be large and average temperature may give misleading values.

TABLE X. DOSE INTERCOMPARISON FOR 10 MeV ELECTRON ACCELERATOR

Date of irradiation: 11-3-98; Date of measurement: 6-4-98				
Irradiation temp.(°C)	Average temp. (°C)	BARC dose(kGy)	RISO dose(kGy)	BARC/RISO
19-27	23	10.7	10.7	1.00
19-35	27	22.3	22.5	0.99
19-43	31	34.6	34.5	1.00
				Average: 0.997

TABLE XI. DOSE INTERCOMPARISON FOR  $^{60}\text{Co}$  GAMMA RAYS

Irradiation temperature: 35.0°C; Date of irradiation: 1-9-98; Date of measurement: 7-10-98

BARC dose(kGy)	RISO dose(kGy)	BARC/RISO
12.0	11.9	1.01
21.4	21.9	0.98
27.6	27.8	0.99
Average : 0.993		

TABLE XII. DOSE INTERCOMPARISON FOR 10 MeV ELECTRON ACCELERATOR

Date of irradiation: 3-9-98; Date of measurement: 8-10-98

Irradiation temp.(°C)	Average temp. (°C)	BARC dose(kGy)	RISO dose(kGy)	BARC/RISO
22–30	26	9.96	9.9	1.01
22–36	29	18.6	18.5	1.01
22–43	33	29.1	29.2	1.00
Average : 1.007				

## 12. CONCLUSIONS

These studies show that dissolution of 20 mg irradiated glutamine in 10 ml aerated aqueous solution containing  $2 \times 10^{-3}$  mol  $\text{dm}^{-3}$  ferrous ions and  $10^{-4}$  mol  $\text{dm}^{-3}$  xylenol orange in 0.033 mol  $\text{dm}^{-3}$  sulphuric acid gives very reliable results. Fresh FX solution should be prepared and used within a few hours for this purpose. FX solution should be allowed to stabilize for about an hour before use. There is no significant post-irradiation fading and normally a correction is not required for this purpose. The plot of absorbance against dose is non-linear, however, a plot of inverse of absorbance versus inverse of dose is linear which is used for dose calculation. There is a change in the slope of this plot around 15 kGy of dose, however, for a calibration around 25–30°C, there are no significant deviations at this dose. The temperature dependence of glutamine dosimeter during the irradiation is somewhat complex. A calibration curve at 25–30°C is used for dose calculation and the absorbance for irradiations at other temperatures is corrected to this temperature. The dose intercomparison results showed that the dose calibrations at BARC are accurate and the glutamine dosimeter can be used with confidence both for photons and electrons of widely varying energies.

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# CHARACTERIZATION AND EVALUATION STUDIES ON SOME JAERI DOSIMETRY SYSTEMS

T. KOJIMA, H. SUNAGA, H. TACHIBANA, H. TAKIZAWA, R. TANAKA  
Japan Atomic Energy Research Institute,  
Takasaki, Japan

## Abstract

Characterization and evaluation studies were carried out on some JAERI dosimetry systems, mainly alanine-ESR, in terms of the influence on the dose response of parameters such as orientation at ESR analysis, and the temperature during irradiation and analysis. Feasibility study for application of these dosimetry systems to electrons with energies lower than 4 MeV and bremsstrahlung (X rays) was also performed parallel to their reliability check through international dose intercomparison.

## 1. INTRODUCTION

JAERI has developed high-dose dosimetry technique for absorbed doses of  $10^2$ – $10^6$  Gy for  $^{60}\text{Co}$  gamma rays and electrons with energies of 0.15–3 MeV on the basis of physical and chemical dosimetry systems. The parallel-plate ionization chamber has been developed as reference dosimetry system for high-dose rate gamma fields, which is periodically checked at Electrotechnical Laboratory, the PSDL of Japan. Alanine dosimetry system employing rod-shape and thin film dosimeters was also developed as a transfer system for gamma rays and electrons, respectively. Rod-shape alanine dosimeters moulded by injection were also developed as routine dosimeters for both radiations.

Several studies mainly on these alanine dosimeters were carried out over these years, aiming to understand and improve their characteristics as reference/transfer dosimeters or routine dosimeters for  $^{60}\text{Co}$  gamma rays, electrons and bremsstrahlung. Reliability check of these dosimetry system was also performed through dose intercomparison with other dosimetry standard laboratories.

## 2. RESULTS AND DISCUSSION

### 2.1. Orientation effect of alanine dosimeters during electron spin resonance (ESR) analysis

Different orientations of an alanine dosimeter in the cavity during ESR analysis may affect the dosimeter response. This effect was studied for five different types of alanine dosimeters, as shown in Table I, with different shapes (rods with different lengths and thin film) and for different moulding procedures (press, extrusion, and injection)[1, 2]. The variation in ESR signal amplitude was measured by rotating a dosimeter around the vertical mid-axis and at right angle to the magnetic field in the ESR cavity, at a temperature of 22°C. The orientation effect for a rod dosimeter with both moulding procedures (press and extrusion), even with different lengths, was negligible; although injection-moulded dosimeters show signal amplitude variation of about  $\pm 9\%$  (see Fig.1). However, the influence of this effect on the injection-moulded rod-shape dosimeters can be reduced to  $\pm 3\%$  or  $<0.2\%$  by averaging of the dose response measured for either two different orientations ( $0, 90^\circ$ ) or three

TABLE I. MAIN PROPERTIES OF FIVE DIFFERENT TYPES OF ALANINE DOSIMETERS

Dosimeter	Moulding method	Dimension (mm)	Density (g/cm <sup>3</sup> )	Composition (weight %)
Rod(A) <sup>a</sup>	Extrusion + press	$\phi$ 2.988±0.02 length 20	1.300	DL- $\alpha$ -alanine 70 Polystyrene 30
Rod(B)	Extrusion	$\phi$ 3.064±0.18 length 20	1.147	DL- $\alpha$ -alanine 50 Polystyrene 50
Rod(C)	Injection	$\phi$ 2.976±0.01 length 20	1.210	DL- $\alpha$ -alanine 50 Polystyrene 50
Film(A)	Press	30×8 0.150±0.020	1.220	DL- $\alpha$ -alanine 60 Polyethylene 40
Film(B)	Extrusion	30×8 0.233±0.015	0.812	DL- $\alpha$ -alanine 60 Polyethylene 40

a — Rod(A) = Aminogray

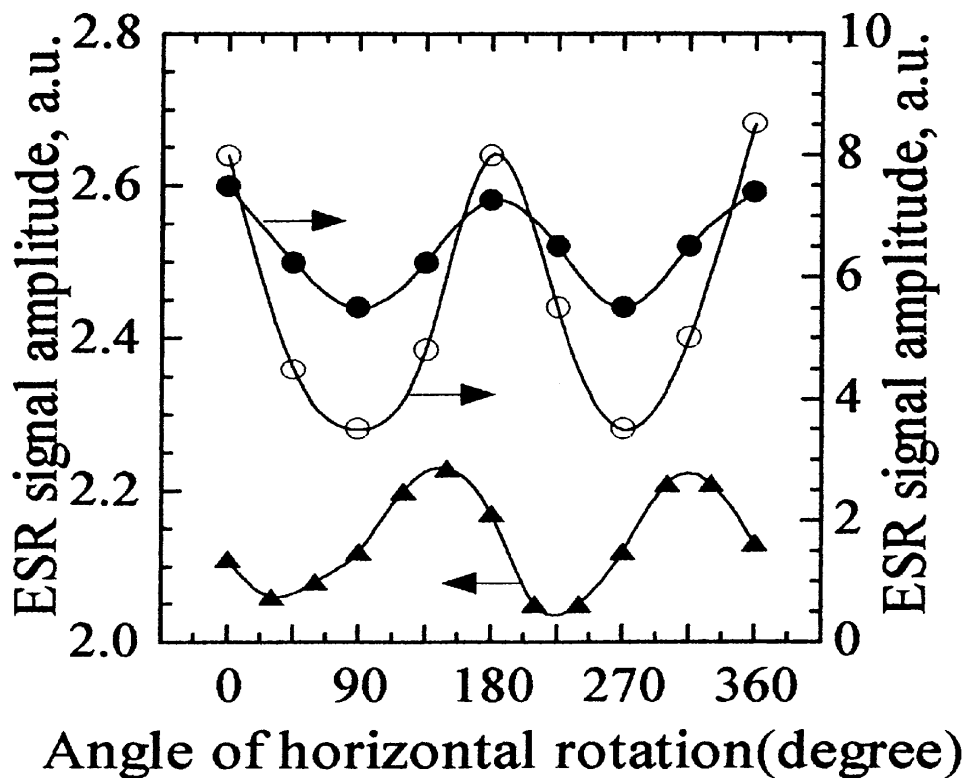


FIG.1. Orientation angular dependence of ESR signal amplitude (peak-to-peak) for film dosimeters moulded by press or extrusion, and for rod dosimeters moulded by injection [1,2].  
o — press-moulding (film), • — extrusion (film), ◆ — injection (rod).

(0, 60, 120°) in the cavity, respectively[2]. Thin-film dosimeters moulded by extrusion also show a pronounced orientation effect compared with press-moulded ones as also shown in Fig.1, when samples are cut into disks and positioned horizontally (parallel to the magnetic field) in the ESR cavity for readout. This effect also can be eliminated when they are positioned vertically and parallel to the mid-axis of the ESR cavity, or positioned vertically in the case of extruded dosimeter, with the custom-made holder consisting of two parallel quartz plates[1].

## 2.2. Irradiation and ESR analysis temperature dependence of alanine dosimeter response

Response characteristics of the alanine-polystyrene (PS) dosimeters, Aminogray, were studied at an absorbed dose of 5 kGy over the low irradiation temperature range  $-196$  to  $30^{\circ}\text{C}$ , and the ESR analysis temperatures in the range of  $0$  to  $50^{\circ}\text{C}$  over the dose range  $0.1$ – $10$  kGy[3]. The irradiation temperature coefficient of  $+0.24\%$  of the response/ $^{\circ}\text{C}$  previously estimated for  $0$ – $70^{\circ}\text{C}$  was verified down to  $-78^{\circ}\text{C}$ , although the value at  $-196^{\circ}\text{C}$  was much different as shown in Fig. 2. This value, however, is in good agreement with the collaborative results obtained at CERN for  $-196$  and  $-269^{\circ}\text{C}$  (4K) at a dose of 1 kGy [4].

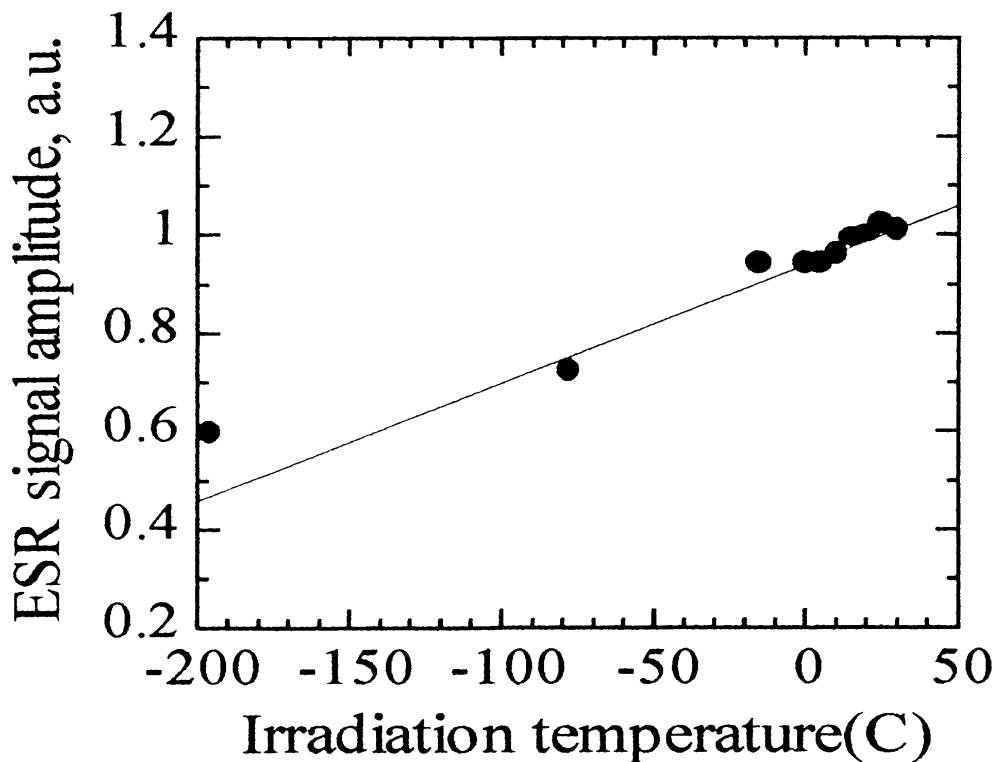


FIG.2. ESR signal amplitude of alanine-PS dosimeters (Aminogray) irradiated to 5 kGy at different irradiation temperatures. Solid line is on the basis of an irradiation temperature coefficient of  $+0.24\%$  of the response/ $^{\circ}\text{C}$ [1].

The ESR intensity decreases with temperature during ESR analysis following the reciprocal of the absolute temperature based on Boltzman's constant. The average value of the ESR-analysis temperature coefficient for alanine-polystyrene dosimeter is -0.25% of the response/°C in the temperature range 0–50°C. This can be eliminated practically by frequent measurements of a reference ESR standard.

### **2.3. Irradiation temperature dependence of PMMA and silver-dichromate dosimeter responses**

Characteristics of undyed/dyed polymethylmethacrylate (PMMA) dosimeters and silver-dichromate dosimeters were studied under collaborative works with OAEP (Thailand) and VAEC (Vietnam) for the improvement of the performance of the existing dosimetry systems [5, 6].

Effects of low irradiation temperature on the gamma ray response were studied for dyed and undyed PMMA dosimeters, Radix RN-15(Batch 4), Red 4034(Batch-D), Amber 3042(Batch-D) and Gammachrome YR(Batch 3) at three different gamma dose rates of 1.0, 4.9 and 6.5 kGy/h for irradiation temperatures from -196 to +45°C and the doses of 2 and 25 kGy. Radix RN15 has nearly linear dependence at 25 kGy with a temperature coefficient of +0.15% of the response/°C and +0.25%/°C for temperatures below and above -15°C, respectively. Red 4034 and Amber 3042 have smaller dependence in this temperature range at 25 kGy, while both show different tendency at -78 and -196°C. Dose response of the Amber dosimeter increases with temperature with a coefficient of +0.5% of the response/°C above -78°C at 2 kGy. The Gammachrome YR response below -78°C is almost 30% higher than that at 20°C and decreases with increasing temperature above -78°C with a coefficient of -0.3% of the response/°C.

Effects of temperature during <sup>60</sup>Co gamma ray irradiation and spectrophotometry analysis on the dose response were studied in the irradiation temperature range 5–60°C for low-dose and high-dose silver-dichromate dosimeters for the dose ranges 2–10 kGy and 10–50 kGy, respectively. The temperature coefficient in the range 25–60°C was estimated to be -0.20% and -0.26% of the response/°C at doses of 2–10 kGy and 10–50 kGy, respectively; although there is a slight tendency of dose dependence. The effect of temperature during spectrophotometry analysis on molar extinction coefficients was not appreciable in the range 5 to 50°C.

### **2.4. Alanine-PS routine dosimeters moulded by injection and its application to electron irradiation**

Preparation procedure of rod-shaped alanine-PS dosimeters was simplified using injection moulding to facilitate their commercialization for large-scale use in industrial facilities as an inexpensive routine dosimeter [2]. This dosimeters (50wt% DL- $\alpha$ -alanine and 50wt% PS) is 3 mm in diameter, 20 mm in length and has mass of  $0.169 \pm 0.0007$ g. The precision of dose-response over the dose range of 0.1–100 kGy is estimated to be within  $\pm 2\%$  ( $2\sigma$ ), without correction for sample-to-sample mass scattering, when taking average of the three measurements with different dosimeter orientation in the ESR cavity as described in Section 2.1. The irradiation temperature coefficient at a dose of 1 kGy is 0.25% of the response/°C in the temperature range 5–45°C. The fading of the radiation-induced ESR signal

at doses of 0.1–100 kGy is less than 1% for at least 2 months after irradiation when stored at 25°C and relative humidity of 40%.

Thin film dosimeters with the thickness less than a few hundreds  $\mu\text{m}$  are commonly used for routine dosimetry of electrons with energies lower than 4 MeV without consideration of the depth dose distribution within the dosimeter. In one-sided irradiation processes, however, the rod-shaped alanine dosimeter has a potential to be used as a dose monitor at a specific position in the product, when ratio between the ‘surface dose’ given by the thin film dosimeter and the ‘average dose’ given by the rod-shaped dosimeter is known. A polyethylene block ( $40 \times 50 \times 10$  mm) with horizontally long holes was used to adjust the centre axis of rod-shaped alanine dosimeters (3 mm in dia.) at 2 mm depth. Alanine dosimeters were irradiated using 1–3 MeV electrons (scan width is 60 cm) together with the 3 mm-thick stack of cellulose-triacetate (CTA) films covering the same range of 0.5–3.5 mm in depth in the block. The ratios of the ‘surface dose’ estimated using the shallowest CTA film and the ‘average dose’ measured by the alanine dosimeter were  $0.83(\pm 4\%)$  for energies above 1.2 MeV, while the ratios of ‘average dose’ in the entire CTA stack and that measured by the alanine dosimeters agreed within  $\pm 2\%$ . Once the ratios are given under actual radiation processing condition, rod-shaped alanine dosimeters can be used for routine dose monitoring for electrons of energies higher than 1.2 MeV.

## **2.5. Establishment of $^{60}\text{Co}$ gamma ray dose calibration laboratory and reliability check of dosimetry through inter-laboratory comparison**

A  $^{60}\text{Co}$  gamma ray dose calibration facility has been developed at JAERI to establish suitable irradiation geometry employing two different plaque sources to provide wide uniform dose-rate irradiation fields covering dose rate range of 5–200 Gy/h and 400–20,000 Gy/h, and an air-conditioning vessel controlling the irradiation temperature in the range 5–90°C ( $\pm 0.3^\circ\text{C}$ ) and relative humidity in the range 30–90% ( $\pm 2\%$ ) [7].

The dosimetry procedure was thoroughly reviewed, and the overall uncertainty in the dose value measured at JAERI was estimated in terms of a combination of random and other uncertainties, to demonstrate the quality of metrological performance [8]. The overall uncertainty in gamma ray dose employing an ion chamber as a reference dosimeter and alanine dosimeters as transfer dosimeters is estimated to be 2.2 and 3.4% at a 95% confidence level, respectively.

Dose intercomparison studies in the dose range 1–50 kGy were carried out two times with National Physical Laboratory (NPL, UK) to confirm our overall uncertainty value, employing NPL and JAERI alanine dosimeters as well as NPL dichromate dosimeters. There is significant difference in irradiation geometry due to different source arrays: a plaque source at JAERI and a cylindrical source at NPL. A PMMA phantom designed for dosimeter accommodation was used for the JAERI irradiation field to establish electron equilibrium and to eliminate electron contamination from surrounding materials. All the results using alanine and dichromate dosimeters show good agreement, within about 2% as shown in Fig. 3, which is consistent with combined value of each overall uncertainties in doses given by NPL and JAERI.

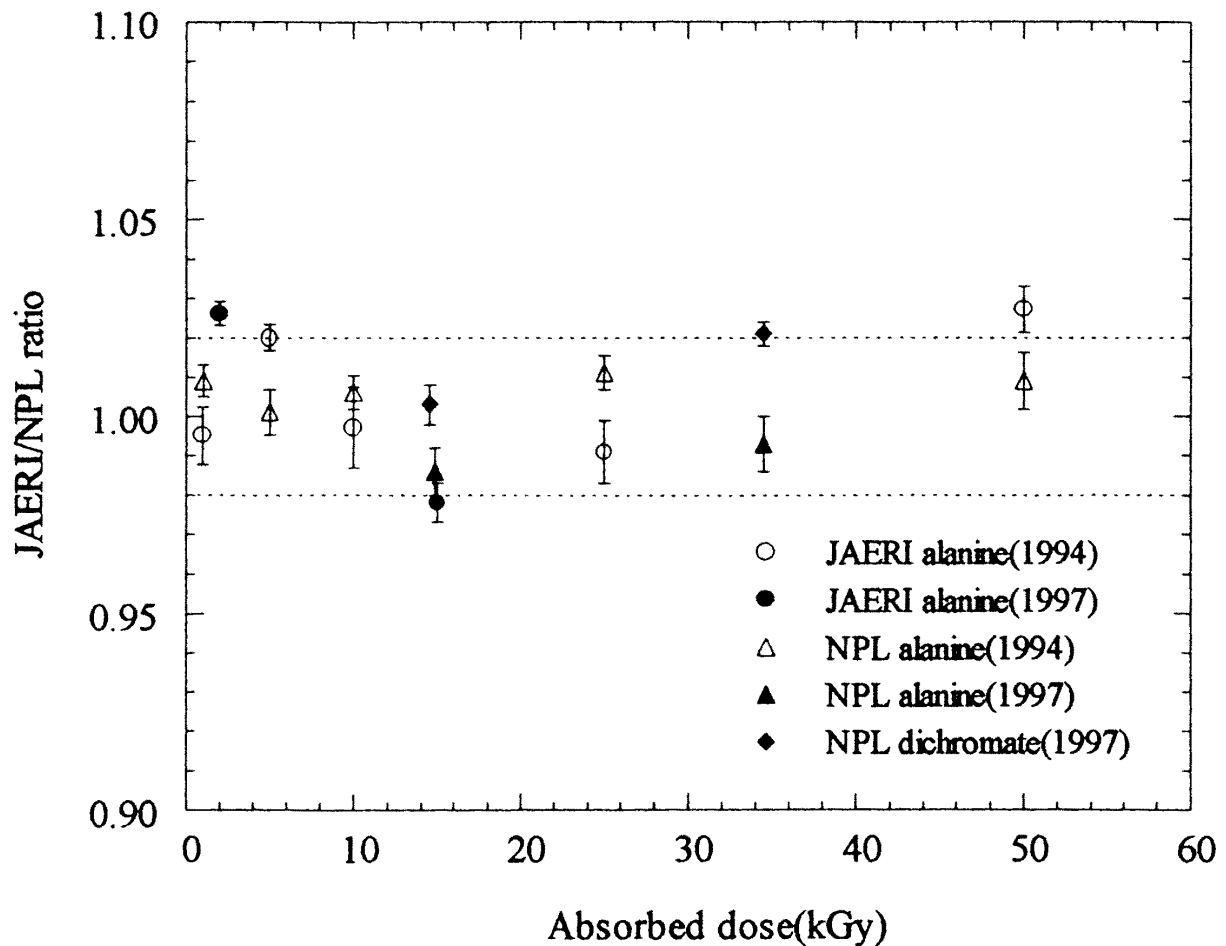


FIG.3. The results of dose intercomparison study performed between NPL and JAERI, employing NPL-alanine, NPL-dichromate and JAERI-alanine dosimeters (Aminogray) as transfer dosimeters [8].

Another dose intercomparison study has started for electron doses involving alanine film dosimeters and CTA film dosimeters. Comparison between gamma ray dose and electron dose is also in progress through this study.

## 2.6. Application of existing dosimetry systems for bremsstrahlung (X rays)

Dose rate dependence characteristics of undyed PMMA (Radix RN-15) and alanine-PS rod dosimeters (Aminogray) were studied covering wider dose rate range for their application to dosimetry for X rays obtained from 3 MeV electrons (current: 20 mA) [9]. The dependence was negligible in the dose rate range 5 to 400 kGy/h (see Fig. 4). These dosimeters were irradiated to 10 kGy, where temperature rise during irradiation was negligible. The ratios of the effective mass energy-absorption coefficients of dosimeter materials, alanine or PMMA to those of water were calculated for X rays obtained by 3, 5 and 7 MeV electrons, by weighing the entire energy spectrum given by the DEX-code. Estimated coefficient ratios of  $\mu_{en}/\rho$  for alanine or PMMA to  $\mu_{en}/\rho$  for water are in the range 0.970–0.972 for three different X ray spectra, which were in good agreement with those for  $^{60}\text{Co}$  gamma rays (1.25 MeV as average), as shown in Table II. This result demonstrates feasibility of application of these

dosimeters to X ray, when they are calibrated using  $^{60}\text{Co}$  gamma rays, even though X rays have a broad photon energy spectrum.

TABLE II. EFFECTIVE MASS ENERGY-ABSORPTION COEFFICIENTS ( $\text{cm}^2\text{g}^{-1}$ ) FOR X RAYS

Material	1.25 MeV(Co-60)	3 MV X rays	5 MV X rays	7 MV X rays
Water	0.0297 (1.000)*	0.0302 (1.000)	0.0282 (1.000)	0.0271 (1.000)
Alanine	0.0288 (0.970)	0.0293 (0.970)	0.0274 (0.972)	0.0263 (0.970)
PMMA	0.0288 (0.970)	0.0293 (0.970)	0.0274 (0.972)	0.0263 (0.970)

\*Values in parentheses are ratios of coefficient of alanine and PMMA to those of water

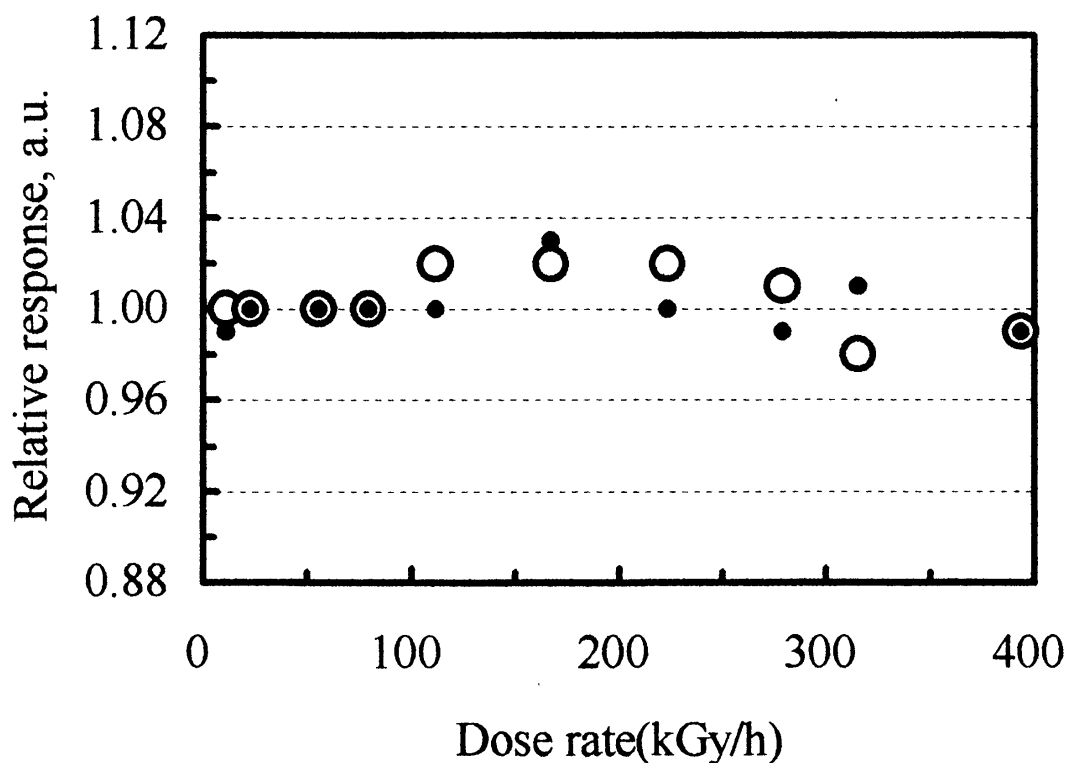


FIG. 4. Dose rate dependence of PMMA and alanine-PS dosimeters for X rays obtained from 3 MeV electrons. ○ — alanine-PS (Aminogray), ● — PMMA (Radix RN-15).

### 3. SUMMARY

Characterization and evaluation studies on some of the JAERI dosimetry systems were performed according to the IAEA co-ordination research project (Research Contract No. 9377). They should contribute to both IAEA and JAERI for establishment/improvement of the new/ existing dosimetry systems for gamma rays, electrons and X rays in high-dose dosimetry for radiation processing. The initiation of the studies on X ray dosimetry is also a key to open a new aspect of radiation processing using high-energy X rays.

## ACKNOWLEDGEMENTS

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# THIN LAYER ALANINE DOSIMETER WITH OPTICAL SPECTROPHOTOMETRIC EVALUATION

Z.P. ZAGÓRSKI

Department of Radiation Chemistry and Technology,  
Institute of Nuclear Chemistry and Technology,  
Warsaw, Poland

## Abstract

Experience in the high dose dosimetry of gamma radiation, gathered in our group from the sixties till now, allows to express the opinion, that techniques applied are adequate to solve problems. It can be confirmed by the fact that 60% of laboratories participating in the international comparison during the duration of the contract obtained satisfactory results. Adaptation of these methods, in particular of the alanine-ESR dosimetry to highly inhomogeneous fields of EB gives poor results, as it has been shown on thin films of the alanine/polymer composite. However, the applications of these films give excellent results if the concentration of the radical  $\text{CH}_3\text{C}\cdot\text{HCO}_2^-$  is measured by diffuse reflection spectrophotometry, which tolerates poor transparency of the composite and is insensitive to the orientation of crystals of alanine in thin films, what is disqualifying the ESR measurements. The development of thin-film dosimeters for EB processing was possible due to new developments in solid state radiation chemistry. The research has revealed some unsolved questions, e.g. of the high temperature coefficient of alanine based dosimeters, of the role of the size of spurs and the necessity to adapt dosimetry to the energy spectrum of electrons, because every type of accelerators differs in that respect.

## 1. INTRODUCTION

The purpose of the research was the development of reliable and cheap high dose dosimeter, useful especially in electron beam radiation processing. The main feature of the latter, i.e. the unit process of irradiation, is a high congestion of isodoses, much higher than it is the case in gamma-irradiation. In gamma radiation processing, even thick dosimeters can be used, with the difference between maximum and minimum dose in limits of  $\pm 1\%$  in the body of dosimeter. Therefore application of liquid chemical systems [1] meets in gamma irradiations no problems, especially because irradiation times are long enough to make natural mixing of the solution by convection efficient. Liquid dosimetric systems are seldom applicable in electron beam processing. Sometimes the reason for that is a particular systems radiation chemistry, which answers to the very high dose rate in another way than it is the case with gammas (e.g. Fricke dosimeter). General reason is in general caused by difficulties in preparation of a thin vessel for liquid dosimetric solution, with controlled thickness of walls. Sometimes liquid systems are unavoidable, mainly in the case of physical dosimetry, i.e. with water calorimeters. The water calorimeter has to be used with special precautions. One has to keep in mind, that the aqueous body of such calorimeter is averaging the dose across the isodose curves. There is no need to remind known facts of relations of the dose in layers of water, of millimetre thickness, placed vertically to the scanned beam. The range of doses can differ by many percentages from the average value, as indicated by the temperature of water in the calorimeter, well mixed mechanically. Several possibilities can be chosen, e.g. the entrance dose equal exit dose; in that case the difference between the maximum dose in the depth of water and the entrance/exit dose is at minimum, but still high from the point of view of dosimetry. Solid state calorimeters allow to lower the difference between the maximum and minimum dose, but are introducing other sources of error.

It is obvious that the dosimeters to be used in electron beam irradiations have to be solid systems, thus avoiding the question of walls, and meeting easily the necessity of constructing

them in any thickness. Acceptation of the thin layer solid systems demands consideration of specifics of solid state radiation chemistry. My group is investigating several aspects of solid state radiation chemistry, especially those, which show differences to radiation chemistry of liquids. Solid state radiation chemistry implies the use of specific measuring methods.

Among solid state dosimeters, the alanine dosimeters with ESR read-out have found a wide acceptance for measuring dose in gamma radiation or bremsstrahlung fields. My group has taken part, during the time of realization of the contract with the IAEA, in inter-laboratory comparisons of results, placing themselves in the group of 60% of laboratories which got acceptable results. One has to stress, that the success of the alanine/ESR dosimetry is limited, at the time being, to gamma and bremsstrahlung radiation fields, i.e. to fields of rather excellent homogeneity, especially in large installations for irradiation, where the object of irradiation is passing places of different dose rate, collecting a well averaged dose of uniform distribution. The alanine dosimeter is prepared by some laboratories in thin films, supposed to be measured in a special, difficult way by ESR but that approach, the only acceptable in EB processing, has never been the subject of inter-laboratory comparisons, for obvious reasons. The IAEA Seibersdorf Laboratory is using its Bruker ESR-Spectrometer for thick alanine dosimeters of Japanese production, having enough difficulties with dealing of bad effects of orientation of alanine crystals in dosimeters, as described by Kojima.

It is obvious that the excellent properties of alanine dosimeter, in which the radical anion formed by deamination:  $\text{CH}_3 \text{C}^\bullet\text{H} \text{CO}_2^-$  has to be measured by another method, not by the ESR with its inherent inconveniences, not mentioning difficulties in reproducible preparation of thin films of a composite containing small alanine crystals. Some unexplained phenomena in a conventional alanine-ESR dosimeters, even in its "thick" shape have appeared early: The ASTM standard for the alanine-ESR dosimeter bears an advice how to make the dosimeter in laboratory, by mixing alanine crystals with paraffin. The recipe mentions conditioning of such dosimeters for several weeks without explanation of the purpose of this procedure and for a mechanism responsible for the phenomenon. Most probably the instability of the alanine/paraffin dosimeter has something to do with the matrix and its relaxation and/or with ESR method of reading out the concentration of the radical anion mentioned above. Another unexplained fact is a very high temperature coefficient of the alanine dosimeter, this time not connected with the ESR method. Our studies on basics of solid state radiation chemistry have found out indications for the solution of the problem.

The only solution of satisfactory measurement of the concentration of the radical was an application of another measuring method, preferably an optical one. Previous efforts on non-ESR measurement, published in the literature were proposing destructive methods unacceptable for the purpose of routine EB dosimetry. Switching to new methods could not be successful without improving our knowledge about the solid state radiation chemistry of alanine in composites.

## 2. SPECIFICS OF SOLID STATE RADIATION CHEMISTRY

As it has been outlined in the 'Introduction', the liquid systems are seldom used in practical electron beam high-dose dosimetry for radiation processing. They involve basics radiation chemistry (e.g. Fricke dosimeter) as fundamental means of connecting the absorbed amount of energy with the measured quantity. Other liquids, like water used as the apparently inert body of the calorimeter also undergo chemical changes in the effect of absorption of energy, but in that case efforts are made to limit the extent of radiolysis, which is responsible

for the heat defect. In that case radiolysis described by radiation chemistry of competing reactions presents a negative phenomenon, from the point of view of dosimetry.

Majority of useful dosimetric systems applied in radiation processing is solid but paradoxically, radiation chemistry of solids is less developed than radiation chemistry of liquids. First feature differing radiation induced chemistry in solids in comparison to liquids is much shorter life of transients in liquid matrix. We have shown [2] that the free radical anion  $\text{CH}_3 \text{C}^\bullet\text{H CO}_2^-$  derived by radiation induced deamination from solid alanine ( $\text{CH}_3 \text{HC NH}_3^+ \text{CO}_2^-$ ) has the life time by 12 orders of magnitude longer than the same radical anion obtained in pulse radiolysis experiment by detachment of hydrogen on  $\alpha$ -carbon of propionic anion, or by detachment of Cl atom from  $\alpha$ -chloropropionate. All three free radical anions are the same species, as they have the same optical absorption spectrum and the same molar absorption coefficients. The outstanding stability of the radical anion  $\text{CH}_3 \text{C}^\bullet\text{H CO}_2^-$ , when formed in the lattice of alanine, is the reason for excellent properties of so called alanine dosimeter, regardless the method of measuring the concentration of the radical — non-damaging methods ESR or DRS (diffuse reflection spectrophotometry) or involving dissolution of irradiated alanine. Our innovation consist in the use of electron absorption spectrum of the radical which has the maximum at 350 nm and exhibits the molecular absorption coefficient  $\epsilon=1100 \text{ M}^{-1}\text{cm}^{-1}$  [3].

Another feature of solid state radiolysis is the phenomenon of spurs. In that case only the primary act is identical in liquid and solid state, but secondary reactions are running differently. The primary act of absorption of ionizing radiation consists, contrary to photochemistry, in interaction with electrons of the medium, irrespectively of their affiliation to a particular molecule. The very first act of interaction may not to be supposed as similar to photochemical, where the electromagnetic quantum is reacting selectively with one type of constituent of the system, e.g. with the solute, in other words with a particular molecule or part of it with the chromophoric group. With high energy quanta it is not the case. Therefore radiolysis starts with the main constituent of the system, i.e. with water, if the object of irradiation is aqueous solution. Another complication is the localization of ionization: Primary ionizing particles, of low LET radiations,  $\gamma$ -quanta or high energy electrons, as well as next generations of quanta and electrons of energies still of the order of keV-s are forming single-ionization spurs, but final generations cause ionizations not very far one from another, called multi-ionization spurs in which products of ionizations, holes and electrons are very close one to another, giving rise to specific reactions.

Single ionization spurs cause the detachment of electron from one small molecule, leaving the positive hole. Multi-ionization spurs cause double and multiple ionizations very close one to another in one molecule what usually leads to the destruction of the molecule with formation of low molecular weight debris. The reactions which follow the primary act of ionization are different for liquid and solid systems in spite of the same chemical composition. The best examples are in the case of water and ice radiolysis and of concentrated liquid aqueous solution as compared with their frozen glasses.

Introduction of pulse radiolysis technique to radiation chemistry of liquids has opened new possibilities for recognition of mechanisms of reactions. Pulse radiolysis of solid state samples is more difficult and seldom applied, but it will be more frequently used in solid state radiation chemistry [4]. Results of pulse radiolysis of single crystals of L- $\alpha$ -alanine are

probing more deeply into the mechanism of deamination of ionized alanine and the formation of the radical anion [5].

More details relevant to the application of solid state radiation chemistry in dosimetry, e.g. the role of dry electron dominating solid state radiation chemistry are published in a general paper [6]. Even more important feature of radiation chemistry of alanine, is described in that paper, i.e. the determination of radiation yields of multi-ionization spurs. These are responsible for 20% of energy deposited by low LET radiation. The occurrence of more than one ionization in one molecule of alanine cause its destruction and not the formation of radical anion mentioned in the Introduction and measured by ESR or DRS. We have determined, by gas chromatographic method the radiation yield of these spurs to amount to 0.95 molecules per 100 eV. That value is increasing with the increase of LET value of the radiation, meaning an increase of the concentration of the debris and causing new phenomena after heavy ions irradiations, previously not understandable.

### 3. METHODS OF MEASUREMENT OF THE RADIOLYSIS PROGRESS AND RESULTS OBTAINED ON CHOSEN DOSIMETRY SYSTEMS

The present work concentrated on the new method of reading the alanine dosimeter, which consists in the formation of radical anion:  $\text{CH}_3 \text{HCNH}_3^+ \text{CO}_2^- \rightarrow \text{CH}_3 \text{C}^\bullet\text{H CO}_2^-$ . The concentration of this radical is usually performed by electron spin resonance method. The ESR signal obtained from irradiated alanine depends on the position of crystals in the resonance cavity of the spectrometer. As the application of single crystals oriented for measurements in the cavity would be impractical and very expensive, the method is applied in irradiation of chaotic, random mixture of microcrystals of alanine. The resulting spectrum is an average of superimposed spectra of single crystals. In the case of thick cylinders of alanine powder, kept in shape by a binder, the homogeneity of the position of crystals is close to ideal, and small inhomogeneities are averaged by recording the signals in several positions of the sample in the cavity, as it is the practice in the IAEA Dosimetry Laboratory.

Turning to thin layers of alanine-polymer composites, the inhomogeneous orientation microcrystals is so bad, that accuracy and precision of measurement becomes catastrophic. In one of our papers [2] we have shown that in the thin layer alanine/polyethylene composite, the orientation of crystals of alanine is already almost complete, so that the ESR signals are more similar to spectra obtained on single crystals properly oriented, but still of mixed character, what makes the precise measurement practically impossible.

We have found that the radical anion  $\text{CH}_3 \text{C}^\bullet\text{H CO}_2^-$  has a well defined optical absorption spectrum in UV ( $\lambda_{\text{max}} = 350 \text{ nm}$ ), with the tail reaching into visible, causing the well known yellowish tint of irradiated alanine. Careful investigation of optical spectra of single crystals, and comparison with ESR spectra has proved the possibility of measuring the concentration of the radical spectrophotometrically in a more convenient way. The spectrophotometric measurement is not sensitive to the orientation of crystals, thus simplifying the measurement even more. There is only one complication, demanding application of diffuse light spectrophotometry for thin films containing alanine. Thin films have to be prepared anyway, therefore the thin film alanine dosimeter together with DRS measurement are creating new approach to electron beam dosimetry.

The influence of multi-ionizations, mentioned in point 2, on the response of alanine dosimeter was investigated and published in [6]. Thermodynamic consideration shows, that double and higher ionization in one molecule of alanine cause its total destruction into debris. One of the products, CO<sub>2</sub> is unique for multi-ionization spurs and cannot be produced in single ionization spurs. It was successfully determined by gas chromatography, so that one can estimate the participation of multi-ionization spurs to 20% of total absorbed energy. This yield in solid state is very similar to low LET radiolysis in aqueous solutions. One has to stress, that debris of alanine decomposition in multi-ionization spurs do not show signals in ESR or DRS spectra. That is another fact explaining the advantage of alanine dosimetry.

Introduction of the DRS method to dosimetry is not limited to thin film alanine composite dosimetry. Many experiments have been made with a thin film composite of sodium chloride crystalline powder with polyethylene. Optical absorption spectra of F-centers in NaCl are again identical with conventional absorption spectra in single crystals, obtained in other laboratories. Proposed dosimeter is very cheap, but its drawback is easier bleaching of the yellow colour than it is the case with alanine dosimeter. The linear signal/dose relationship observed up to 60 kGy with pure crystalline powder is not repeated, for unknown reasons, in the case of the composite. Probably there is some influence of the matrix, not explained yet. The role of matrix in the better known case of alanine is also not quite clear yet [7]. The drawback of the NaCl connected with the calibration curve will be cleared in next investigations; in the mean time only a short note is in print [8].

Another new field of the application of the DRS method to dosimetry is the application to quantification of colour changes in labels used qualitatively or semi-quantitatively as indicators in radiation processing. The investigation is not published yet, except a short note [9]. Full version is considered for publication by 'Radiation Physics and Chemistry', because it was exhibited as a poster during the 11th International Meeting on Radiation Processing (IMRP-11) in Melbourne in March 1999. Although that investigation is not credited to the IAEA, it has been created by inspiration from the IAEA contract.

#### 4. ISODOSE MAPPING IN ELECTRON BEAM PROCESSING

As mentioned in the introduction, the electron beam processing is characterized by the unfavourable congestion of isodoses, much more inconvenient than in gamma irradiations, especially in large installations, where averaging of doses in moving boxes takes place. There is no such possibility in electron beam irradiation, except the sometimes advised double side irradiation. That procedure is very risky, because the slightest change in geometry of filling the material into boxes can cause severe under- or over-exposures of the irradiated material and provoke unfavourable distribution of electric charge [10, 11].

Thin layer dosimeters only can secure the mapping of isodoses in electron beam irradiations. Results are shown in the paper by Zagórski and Rafalski [12]. The isodoses obtained in a flat object irradiated from above with scanned beam of electrons show immediately what one can expect after inserting the object of known size and of density close to the density of the material for which the isodose chart has been prepared. Alanine dosimeters produced for measurements in gamma fields are evidently too thick to obtain proper results because covering many layers of different dose rate.

## 5. CONCLUSIONS

An useful, thin alanine/ESR dosimeter available for routine EB dosimetry is not existing yet, therefore no comparisons can be made. The thin film alanine/DRS dosimetry is at the very beginnings and the efforts put into its developments until now constitute hardly 1% of efforts invested into the alanine/ESR dosimeter. Nevertheless, several points can be formulated already:

- i) The spectrophotometric equipment for the read-out is by one order of magnitude cheaper than the ESR equipment, even of simplified construction which is not completely satisfactory in comparison to the universal ESR spectrometer. The spectrophotometric equipment may be used for other dosimetric measurements, e.g. cellulose acetate films etc. The temperature of measurement is not critical for precision.
- ii) The time needed for measurement of the dose by alanine/DRS dosimeter is shorter in comparison to alanine/ESR dosimeter which demands measurement at several positions of the dosimeter in the cavity.
- iii) The precision of the alanine/DRS dosimeter is comparable to the precision of the alanine/ESR dosimeter and there is no problem to have results in the  $\pm 2\%$  limits. There are common factors, to the ESR and DRS methods, which could influence the accuracy. The knowledge of the temperature of irradiation is needed as in the case of alanine/ESR dosimetry, because the formation of the radical by deamination runs in the same way in both types of the dosimeter. The high dose rate in the EB processing is influencing all types of dosimeters.
- iv) The stability of the radical in both types of dosimeters is of course the same. The spectrophotometric measurement is too short to cause any bleaching of the radical absorption. The photochemical yield of bleaching at the maximum of absorption is very low (quantum yield is lower than 0.001).

Collaboration of my group with the International Atomic Energy Agency, particularly with K. Mehta, contributed to the revision of methods on every level of dosimetric procedure, from the point of view of accuracy and precision. Inter-laboratory dosimetric comparisons (CRP on Characterization and Evaluation of High-Dose Dosimetry Techniques for Quality Assurance in Radiation Processing) performed by the Agency in July-August 1997 have shown that results of my group were satisfactory and proved the veracity of our work. Unfortunately, because of lack of time and insufficient progress in EB dosimetry, the inter-laboratory comparisons were limited to gamma irradiations only.

Our efforts in preparing thin film alanine dosimeters stimulated other participants to look for new methods of making such films (e.g. the blow out method of preparation of films). Our technology got the status of Polish Patent, No. 172237. We are ready to supply anybody with trial batches of the dosimeter and we are starting to prepare the draft of a standard, because the existing ASTM standard for alanine dosimetry does not take into account thin film alanine dosimetry and alternative methods of measurement of concentration of the radical, like diffuse light spectrophotometry (DRS). We are ready to act as experts in the dissemination of knowledge about improved methods of dosimetry in electron beam radiation processing.

As it is the case with every research and development, results are suggesting new problems to be solved. Research on alanine dosimetry has revealed rather high temperature coefficient of the formation of the radical anion, unusual for free radical formation. High temperature coefficient involved into the radical formation causes complications in preparation calibration curves for high doses. There are differences between gamma and electron beam calibrations, due to adiabatic heating in the latter case. Our experiments into that problem are in preparation for publication in collaboration under an European project [13].

Diffuse reflected light spectrophotometry (DRS) has shown its advantages in solid state radiation research, because it accepts almost opaque samples and in double beam measurements shows only species formed by radiation, without involvement of the texture and other properties of the sample. Its importance reaches far from alanine dosimetry.

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# INVESTIGATION OF THE EFFECT OF TEMPERATURE, DOSE RATE AND SHORT-TERM POST-IRRADIATION CHANGE ON THE RESPONSE OF VARIOUS TYPES OF DOSIMETERS TO COBALT-60 GAMMA RADIATION FOR QUALITY ASSURANCE IN THAILAND

S. BIRAMONTRI  
High-dose Calibration Laboratory,  
Radiation Measurement Division,  
Office of Atomic Energy for Peace,  
Bangkok, Thailand

## Abstract

The influences of combined effect of irradiation temperature from  $-80^{\circ}\text{C}$  to  $60^{\circ}\text{C}$  and dose rate between 0.2 and 4 Gy/s on the gamma ray response of several commercial routine dosimeters (Harwell Red 4034, Gammachrome YR, FWT-60-00 radiochromic films, FWT-70-40 optical waveguides, GafChromic films, and Fuji CTA-FTR-125 films) were investigated for quality assurance in radiation processes. Besides, the studies of short term post-irradiation stability for the period of 2 h to 7 days are also presented. The overall results indicate the need for a calibration protocol under conditions of use.

## 1. INTRODUCTION

In Thailand the use of gamma facilities for radiation processing is a growing industry and an active developing technology. Since 1984 the first commercial-scale gamma irradiation facility has been operating for the purpose of sterilization of medical products. In these facilities various types of commercial dosimeters have been used for absorbed dose measurement, which are first developed in laboratory, then transferred to an industrial application and finally to routine process control. The irradiation processes have occurred under the influences of external factors and specific condition due to the irradiation purpose. There are three different ways to calibrate commercial dosimeters used for gamma processing [1]. One way calls for the dosimeters to be irradiated in a calibration facility. The second way calls for the dosimeters to be irradiated in an in-house calibration facility that has an absorbed-dose rate measured by reference or transfer standard dosimeters. The third way calls for the dosimeters to be irradiated together with reference or transfer standard dosimeters in the production irradiator.

In practice the commercial dosimeters are irradiated at calibration facility using high dose rate irradiator at a different location and sending to the industrial irradiator site for analysis, whilst the dosimeters using in commercial facilities are irradiated in a poor defined radiation field over a period of several days and read out some hours after irradiation. This method has the advantage that the dosimeters are irradiated to accurately known absorbed doses under well-controlled and documented conditions and can reduce uncertainty introduced by the readout instrumentation. But in this case the sources of error due to dose rate effect and irradiation temperature include time-dependent instability in dosimeters response after irradiation have to be investigated. Many papers have been published about the environmental effects on the response of Harwell PMMA and FWT-60-00 radiochromic film dosimeters [2–9]. The collaborated work was set up between OAEF and JAERI for investigation the irradiation temperature effect from  $-190^{\circ}\text{C}$  up to  $45^{\circ}\text{C}$  on the response of undyed and dyed PMMA dosimeters at specific dose of 2 and 25 kGy [10]. The present study is aimed to

investigate the combination of two effects of irradiation temperature from  $-80^{\circ}\text{C}$  up to  $60^{\circ}\text{C}$  and dose rate from 0.07 up to 4 Gy/s and short term post irradiation change on the response of several types of commercial dosimeters.

## 2. EXPERIMENTAL AND RESULTS

Various commercial types of dosimeters were used for this study. The characteristics of these dosimeter systems are list in Table I.

TABLE I. CHARACTERISTICS OF DOSIMETRY SYSTEMS

Dosimeters Type (nm)	Commercial name (kGy)	Nominal size	Method of Analysis wavelength	Analysis dose range	Useful dose range	Supplier (mm)
Polymethyl-methacrylate PMMA	Red 4034 Gammachrome YR	$30 \times 11 \times 3$ $30 \times 11 \times 1.5$	vis. spectrophotometer	640	5-50	AEA Technology, Harwell Laboratory United Kingdom
Radiochromic Film DM-1260	FWT-60-00 Gafchromic	$10 \times 10 \times 0.05$ roll by 10 wide	vis. spectrophotometer	510,605	1-50	Far West Technology Inc., Goleta, USA
Radiochromic Optical Wave-guide	FWT-70-40m	3 dia $\times$ 50 long	photometry	600,656	0.01-1	Far West Technology Inc., Goleta, USA
Cellulose triacetate film(CTA)	FTR -125	roll by 8 wide 0.125 thick	UV/vis spectrophotometer	280	5-300	Fuji Photo film Minato-ku, Tokyo, Japan

### 2.1. Gammachrome YR polymethylmethacrylate dosimeter [15]

#### 2.1.1. Combined effects of irradiation temperature and dose rate studies.

Harwell Gammachrome YR batch 5 are commercially available sealed in the puncture-resistance polyester/aluminium/polyethylene laminated individual sachets for protection against changing environmental condition specially humidity. The irradiations were carried out using two  $^{60}\text{Co}$  sources, gammacell 220 at absorbed dose rate of 2.7 Gy/s and Gammabeam 650 panoramic source at 0.18 Gy/s. The dosimeters were conditioned for 1 hour and irradiated at each temperature using a double-walled glass dewar flask and stored at room temperature immediately after the end of irradiation. The Gammachrome YR were irradiated for the dose range of 100 Gy up to 3 kGy at the following temperature;  $-78$ ,  $-18$ , 0, 25 and  $60^{\circ}\text{C}$ . Fluctuation of control of irradiation temperature was within  $3^{\circ}\text{C}$ . Table II shows the condition in dewar flask at each irradiation temperature. The irradiated dosimeters were kept at about  $25^{\circ}\text{C}$  for 2 h before spectrophotometric (Shimadzu UV-3101PC) analysis.

TABLE II. COOLANTS FOR LOW TEMPERATURE CONDITIONS

Condition in the dewar flask	Temperature( $^{\circ}\text{C}$ )
Ethanol and dry ice	$-78$
Sodium chloride solution and ice	$-18$
Water and ice	0
Water	$+25$

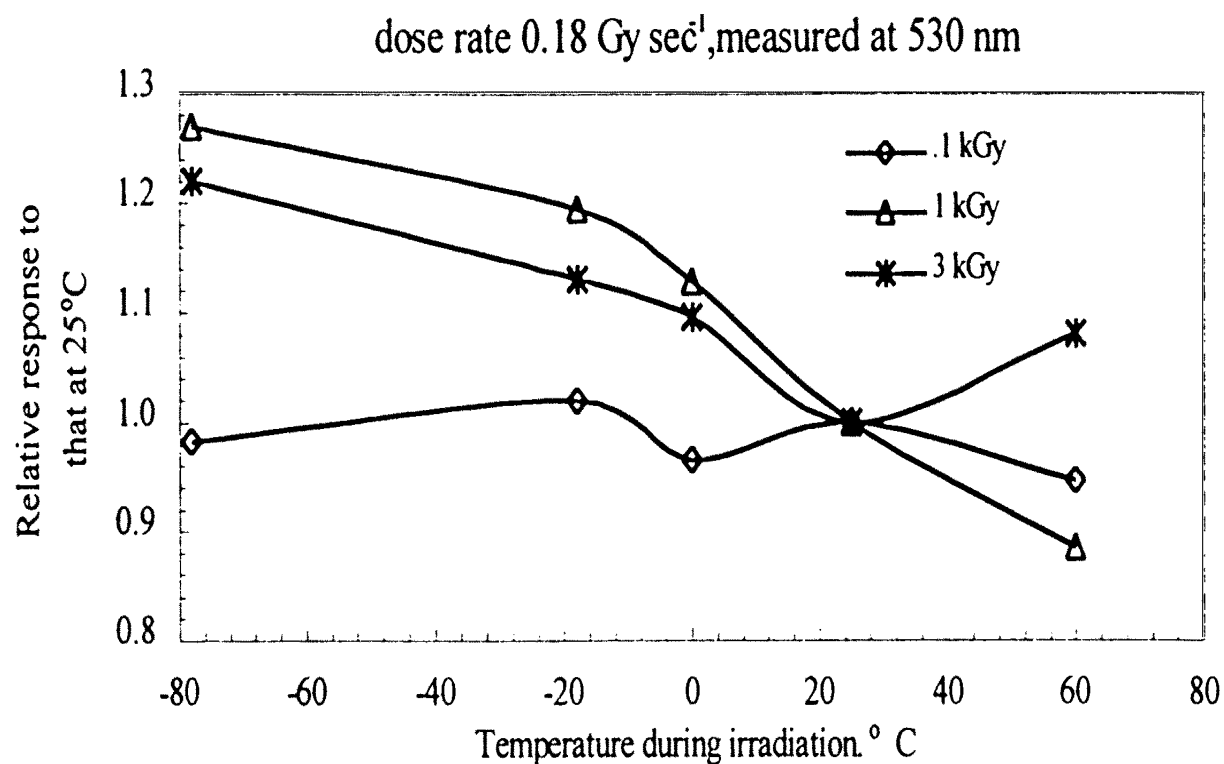
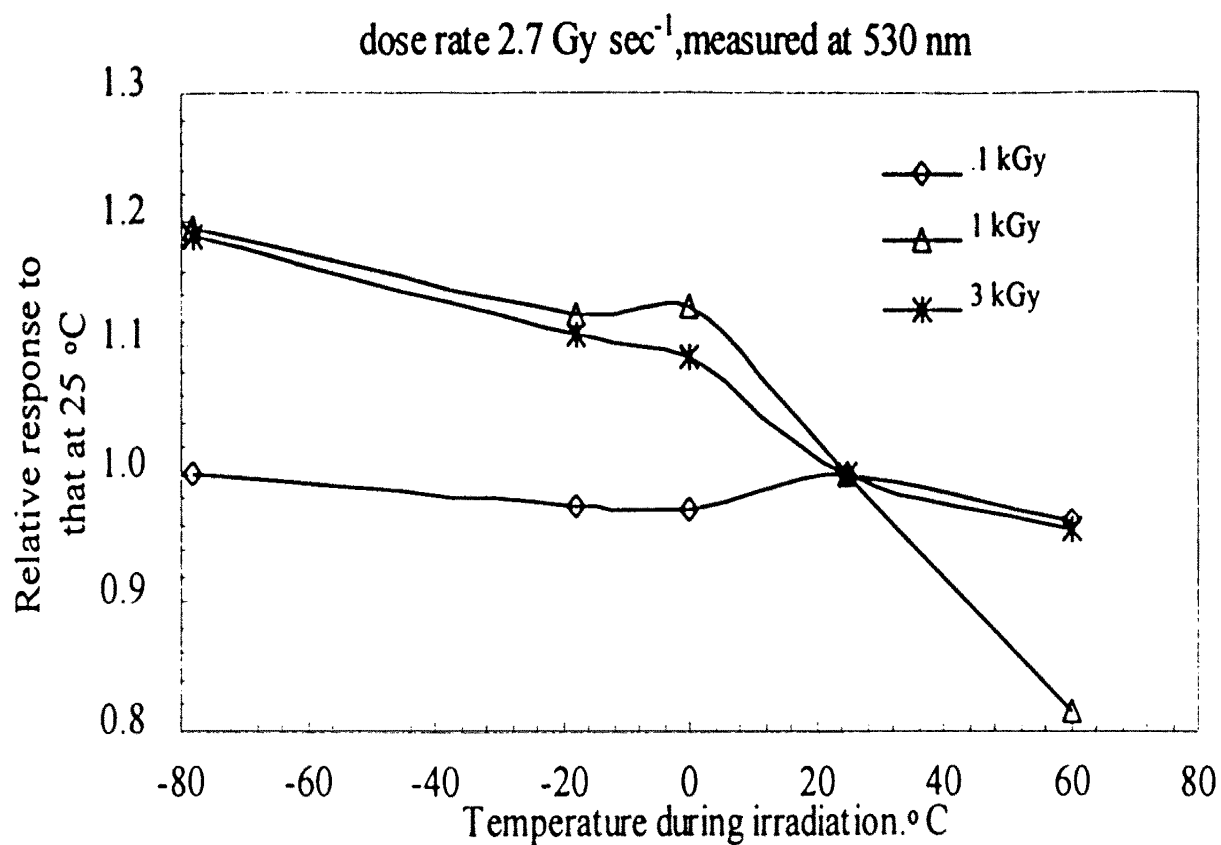


FIG. 1. Temperature dependence, relative to that at 25°C, of gamma response of Gammachrome YR batch 5 at dose rate 2.7 and 0.18 Gy sec<sup>-1</sup>. Where the absorbance per unit thickness was measured at 530 nm.

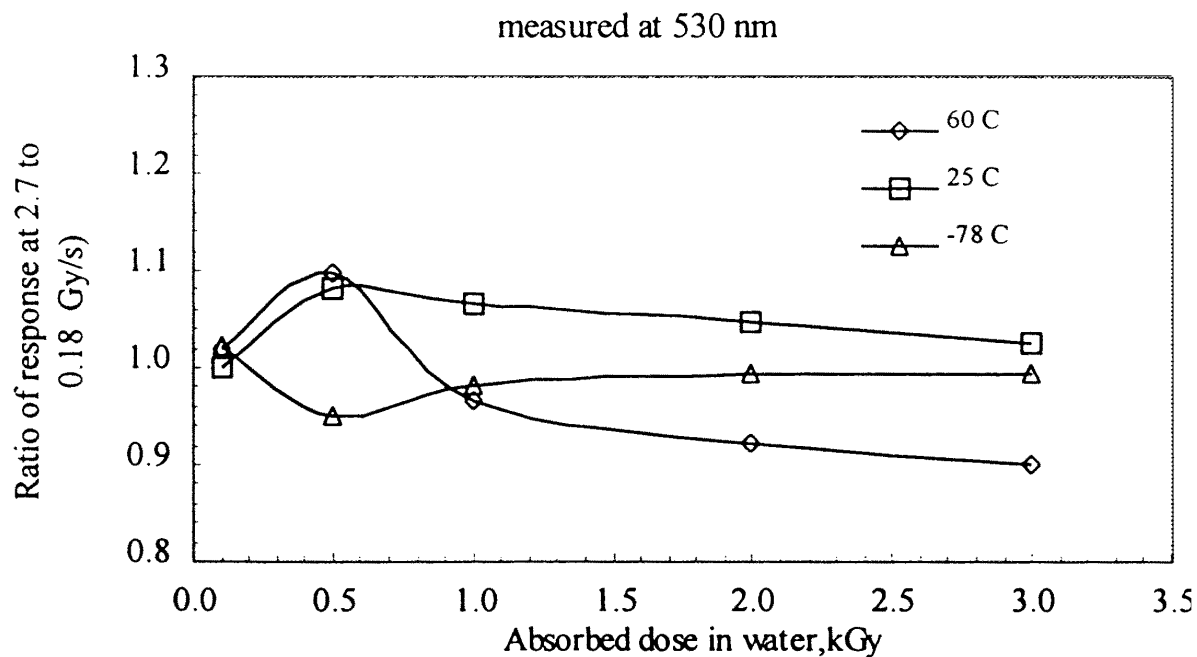


FIG. 2. Combined effect on response of Gammachrome YR batch 5 at does rate of 2.7 and  $0.18 \text{ Gy sec}^{-1}$  with the irradiation temperature: 60.25 and  $-78^\circ\text{C}$ .

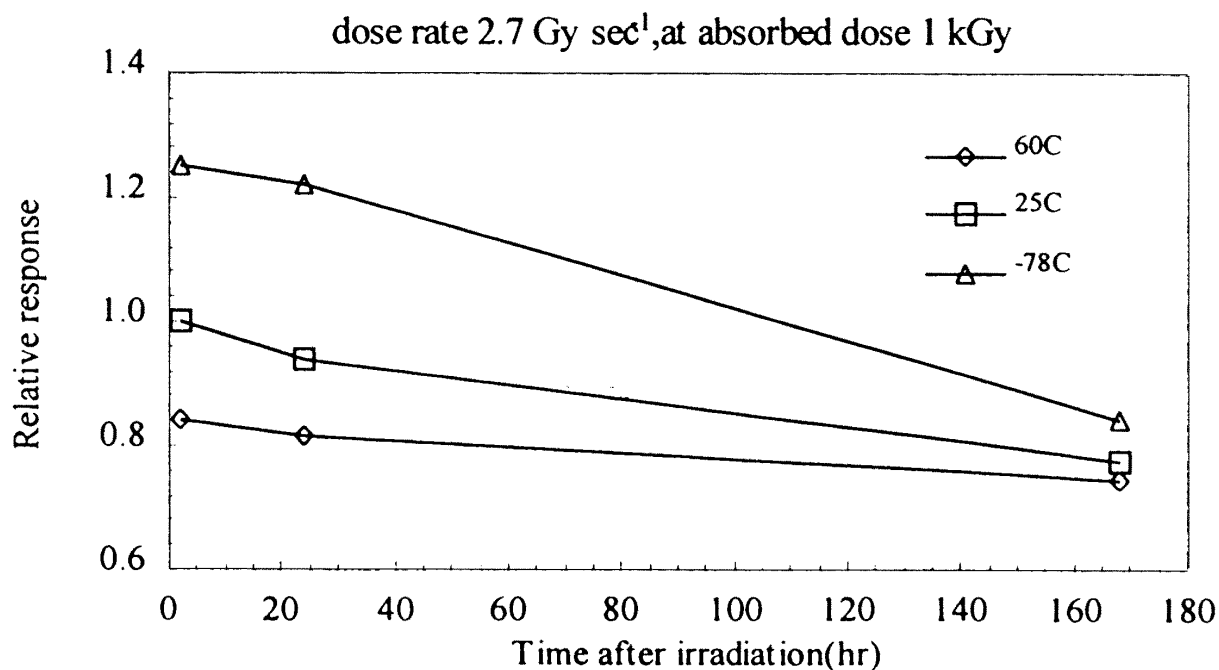


FIG. 3. Post-irradiation studies of Gammachrome YR batch 5 at 1 kGy, analysis time 2.24 hour and 7 days after irradiation for the dose rate  $2.7 \text{ Gy sec}^{-1}$ .

### *2.1.2. Short term post-irradiation studies*

In order to investigate post-irradiation effect nine dosimeters were irradiated at each irradiation temperature for absorbed dose of 1 kGy with the same previous conditions. The irradiated dosimeters were kept in seal before spectrophotometric analysis. The three dosimeters were opened and measured after 2 h whilst the others were measured at 24 h and 7 d after the end of the irradiation.

### *2.1.3. Results and conclusion*

The temperature dependence, relative to that at 25°C of gamma response of GammaChrome YR at dose rate of 2.7 and 0.18 Gy/s for the absorbed dose of 0.1, 1 and 3 kGy are shown in Fig 1. Figure 2 shows the combined effect in term of ratio of response at 2.7 to 0.18 Gy/s at irradiation temperature of 60, 25 and -78°C. The result of the short term post-irradiation studies are shown in Fig 3.

The overall results show only slightly variations in the response of PMMA GammaChrome YR dosimeters for different gamma dose rates in the range from 0.18 to 2.7 Gy/s for irradiation temperatures from -80°C to 60°C.

## **2.2. Radiochromic optical waveguide FWT-70-40m [11]**

Radiochromic optical waveguide is a specially prepared optical waveguide containing ingredients that undergo an ionizing radiation-induced change in photometric absorbance. This change in absorbance can be related to absorbed dose in water. FWT-70-40m are commercially supplied in 3 mm dia × 5 cm long by Far West Technology, Inc. for low dose radiation processing. The dosimeters use hexa(hydroxyethyl) aminotriphenylactonitril dye inside an optical waveguide. Readout of the dosimeter is by photometric means.

### *2.2.1. Combined effects of irradiation temperature and dose rate and short term post-irradiation studies*

The irradiations were carried out using <sup>60</sup>Co Gammabeam 650 panoramic source at dose rate of 3.96 and 0.26 Gy/s. The pre-irradiation absorbance for each dosimeter was read using Far West Opti-chromic photometer reader at 600 and 656 nm. Three dosimeters in 3 mm thick polystyrene tube for electron equilibrium were conditioned for 2 h and irradiated at each temperature using a double-wall glass dewar flask and stored at room temperature immediately after the end of the irradiation. The dosimeters were irradiated for the dose range of 0.02, 0.10, 0.25, 0.5 and 1 kGy at the following temperature: -78, -18, 0, 25, 35 and 60°C using the coolants as shown in Table II. Fluctuation for control of irradiation temperature was within 3°C. The irradiated dosimeters were kept at 25°C for 2 h before absorbance reading and reread again at 24 h and 7 d after irradiation for short term post-irradiation studies.

### *2.2.2. Results and conclusion*

The temperature dependence, relative to that at 25°C, of gamma response of FWT-70-40m batch 4-5 at irradiation temperature of 60, 35, 25, 0, -18, -78°C for two dose rate are shown in Fig 4. Where the absorbance were measured at 600 and 656 nm. The results show only slight variation in the response of dosimeter for different gamma dose rates in the range

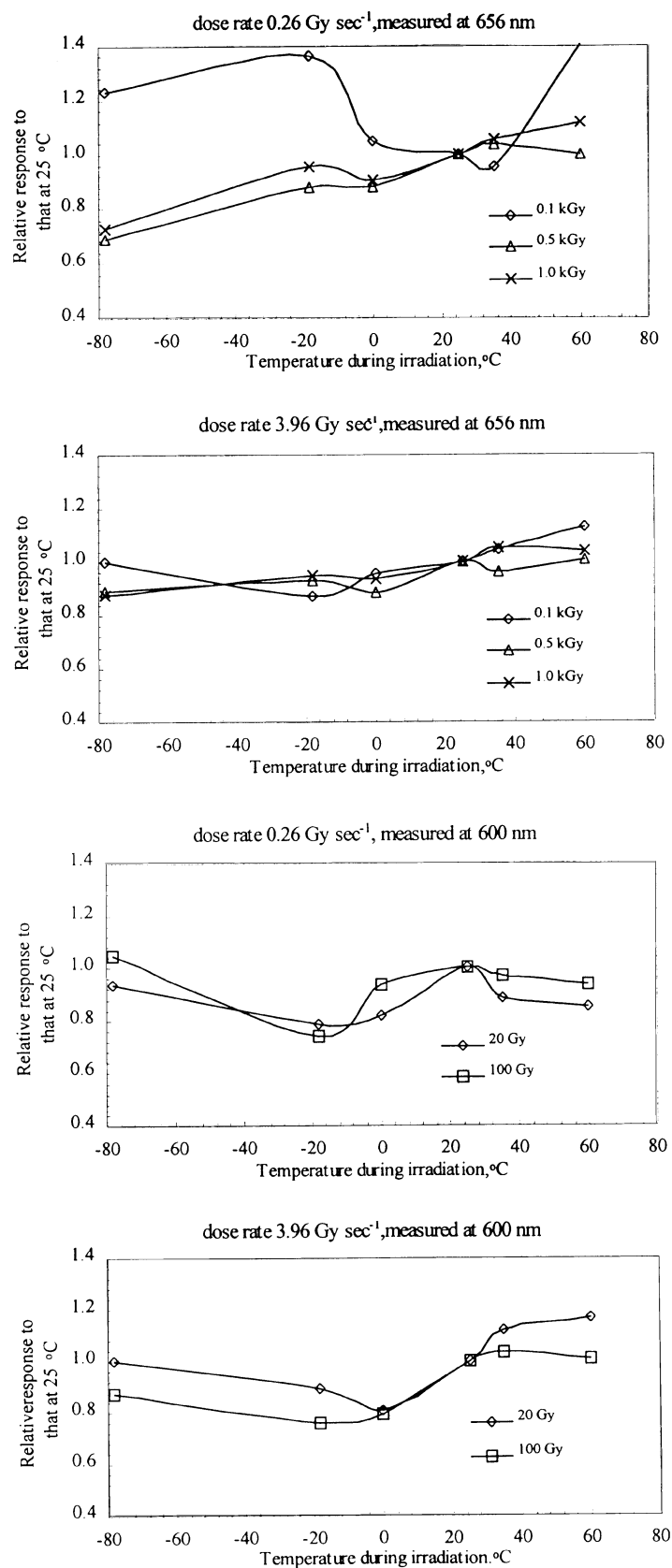


FIG. 4. Temperature dependence, relative to that 25°C, of gamma response of FWT-70-40 m batch 4-5 were irradiated at dose rate 0.26 and 3.96 Gy sec<sup>-1</sup>, measured at 656 and 600 nm (temperature during irradiation: -78, -18, 0, 25, 35, 60°C).

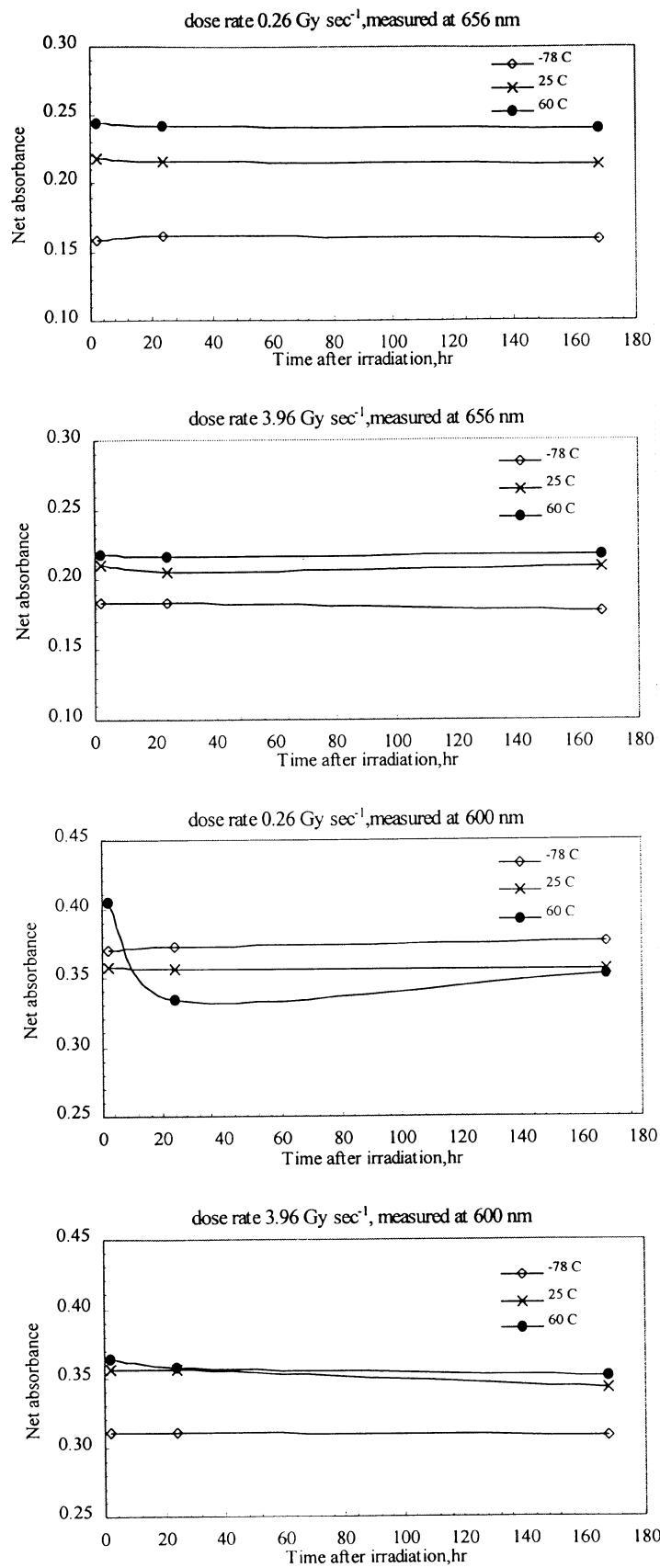


FIG. 5. Short term post-irradiation studies on the gamma response of FWT-70-40 m batch 4-5 at 1 kGy, analysis time 2.24 hr and 7 d after irradiation at dose rate 0.26 and 3.96 Gy sec<sup>-1</sup>, measured at 656 and 600 nm (temperature during irradiation; -78, 25, 60°C).

from 0.26 to 3.96 Gy/s for irradiation temperature  $-18^{\circ}\text{C}$  to  $35^{\circ}\text{C}$ . The response of irradiated dosimeters show stable over 7 day after irradiation as in Fig 5.

### **2.3. GafChromic<sup>TM</sup> dosimeter media DM-1260**

The transparent radiochromic film, GafChromic dosimeter media, are commercially supplied by Far West Technology, Inc. The dosimeter consists of a  $7\ \mu\text{m}$  thick radiation sensitive layer coated on a  $100\ \mu\text{m}$  thick polyester base [12]. They are colourless, grainless and transparent before exposure to radiation and develop a deep blue colour after irradiation. GafChromic<sup>TM</sup> DM-1260 is available in a roll of film 10 mm wide.

#### *2.3.1. Combined effects of irradiation temperature and dose rate and short term post-irradiation studies*

The GafChromic film were cut into  $10\times 10\ \text{mm}$  for fitting to the standard 10-mm cuvette holder of spectrophotometer. Three films for each irradiation were held between two electron equilibrium layer of 3-mm thick acrylic and sealed at 60% r.h. in the polyethylene-aluminium foil laminate for protection against changing environmental condition specially humidity. The pre-irradiation absorbance for each dosimeter was read using Shimadzu UV-3101PC UV/vis spectrophotometer at 400, 500 and 580 nm. The irradiation were carried out using  $^{60}\text{Co}$  Gammabeam 650 panoramic source at 0.26 and 3.96 Gy/s for dose range 0.02 to 1 kGy. The dosimeters were conditioned for 2 h at each irradiation temperature before irradiation and then irradiated using a double-walled glass dewar flask for temperature control. The dosimeters were irradiated for the dose range of 0.02, 0.1, 0.35, 0.5 and 1 kGy at the following temperatures:  $-78$ ,  $-18$ , 0, 25, 35 and  $60^{\circ}\text{C}$  using the coolants as shown in Table II. Fluctuation for control of irradiation temperature was within  $3^{\circ}\text{C}$ . The irradiated dosimeters were kept at  $25^{\circ}\text{C}$  for 2 h before absorbance reading and reread again at 24 h and 7 d after irradiation for short term post-irradiation studies.

#### *2.3.2. Results and conclusion*

The temperature dependence, relative to that at  $25^{\circ}\text{C}$ , of GafChromic DM-1260 at dose rate of 0.26 and 3.96 Gy/s for the dose range from 0.02 to 1 kGy over the irradiation temperature  $-78^{\circ}\text{C}$  to  $60^{\circ}\text{C}$  are shown in Fig 6, where the absorbance per unit thickness were measured at 580 nm. The post-irradiation studies in Fig 8 show very slightly variation for irradiation temperature over  $-78^{\circ}\text{C}$ . Figure 7 shows significant dose rate effects.

### **2.4. Cellulose acetate dosimeter(CTA) [13]**

#### *2.4.1 Combined effects of irradiation temperature and dose rate and short term post-irradiation studies*

The untinted cellulose triacetate(CTA) FTR-125 film dosimeters are commercially supplied by Fuji Photo Film Co., Tokyo, Japan. The dosimeters were cut into  $8 \times 10\ \text{mm}$  for fitting to the standard 10 mm cuvette holder of spectrophotometer. Three films for each irradiation were held between two electron equilibrium layer of 3-mm thick acrylic and sealed at 60% r.h. in the polyethylene-aluminium foil laminate for protection against changing environmental condition specially humidity. The pre-irradiation absorbance for each dosimeter was read using Shimadzu UV-3101PC UV/vis spectrophotometer at 280 nm. The



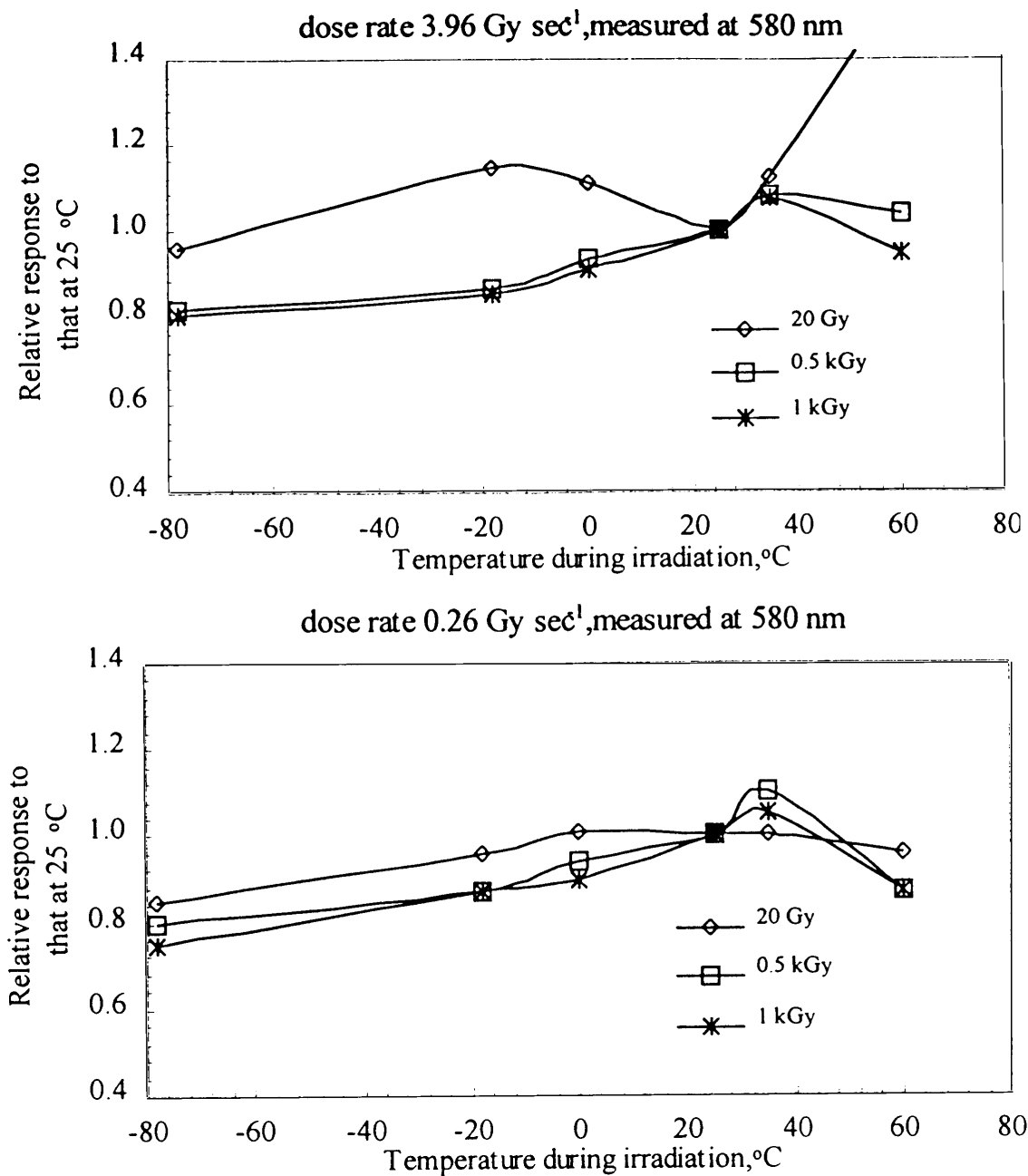


FIG. 6. Temperature dependence, relative to that at 25°C, of gamma response of GafChromic at dose rate 3.96 and 0.26 Gy sec<sup>-1</sup>. Where the absorbance per unit thickness was measured at 580 nm.

irradiations were carried out using <sup>60</sup>Co gammacell 220 at dose rate of 2.26 and 0.27 Gy/s. The dosimeters were irradiated at 5, 10, 15, 20 and 25 kGy at the following temperatures: -18, 0, 25, 35 and 60°C using TURBO-JET air compressor control. Fluctuation for control of irradiation temperature was within 1°C. The irradiated dosimeters were kept at 25°C for 2 h before absorbance reading and reread again at 24 h and 7 d after irradiation for short term post-irradiation studies.

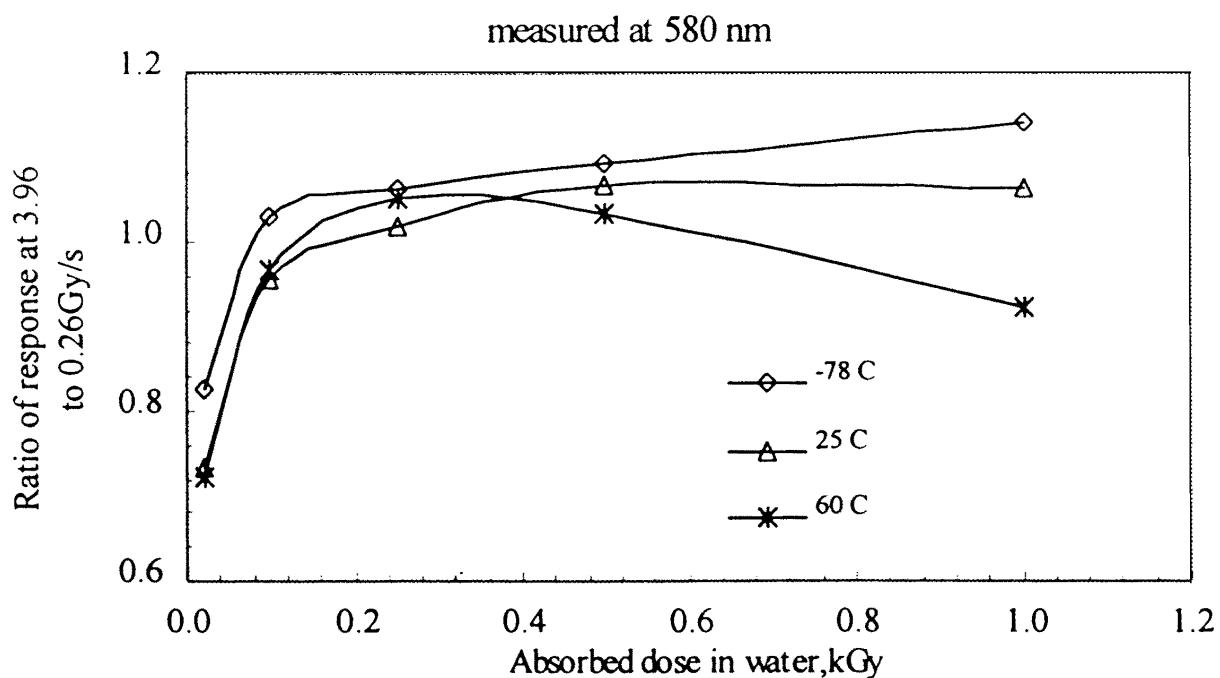


FIG. 7. Combined effect on the response of GafChromic at dose rate of 3.96 and 0.26 Gy sec<sup>-1</sup> with the irradiation temperature of 60, 25 and -78°C.

#### 2.4.2. Results and conclusion

The effect of irradiation temperature on the gamma response of CTA over the range-18°C up to 60°C for the dose range of 5 to 25 kGy are shown in Fig 9. Figure 10 shows significant dose rate effect at 2.26 and 0.27 Gy/s for the irradiation temperature of 25 and 35°C. The post-irradiation behaviour of dosimeters cover the range from 2 h to 7 day after irradiation was found slightly variation in Fig 11.

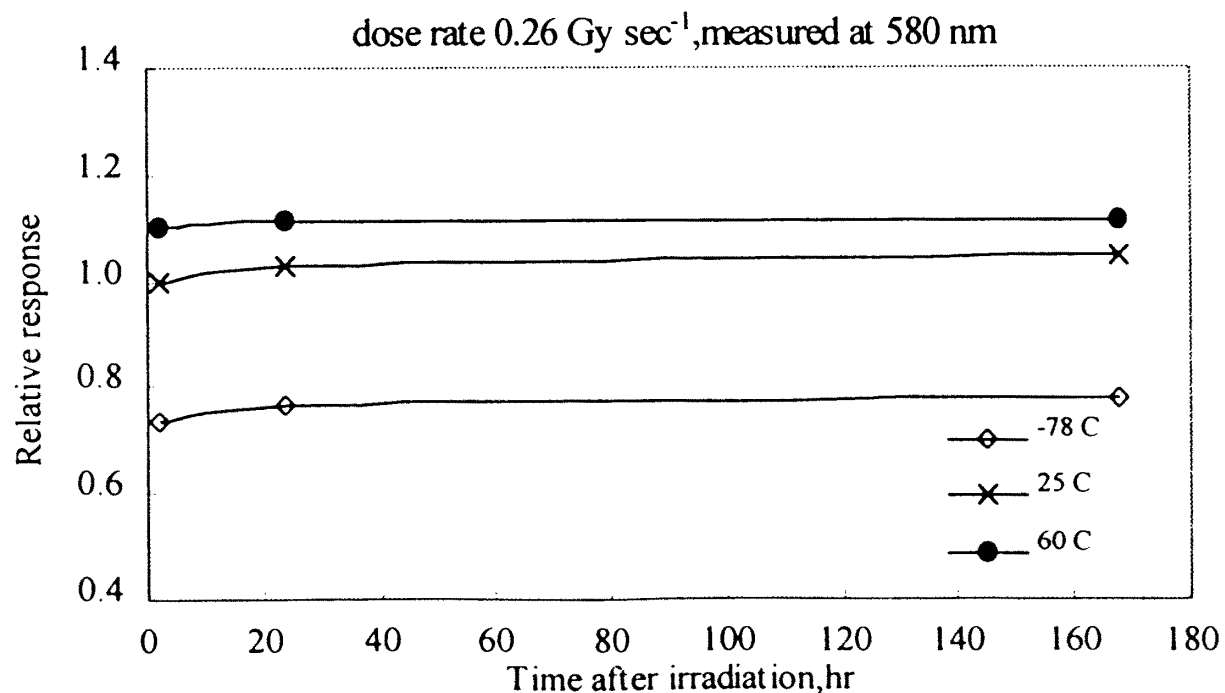


FIG. 8. Short term post-irradiation studies of GafChromic at 1 kGy, analysis time 2.24 hour and 7 days after irradiation for the dose rate of 0.26 Gy sec<sup>-1</sup>.

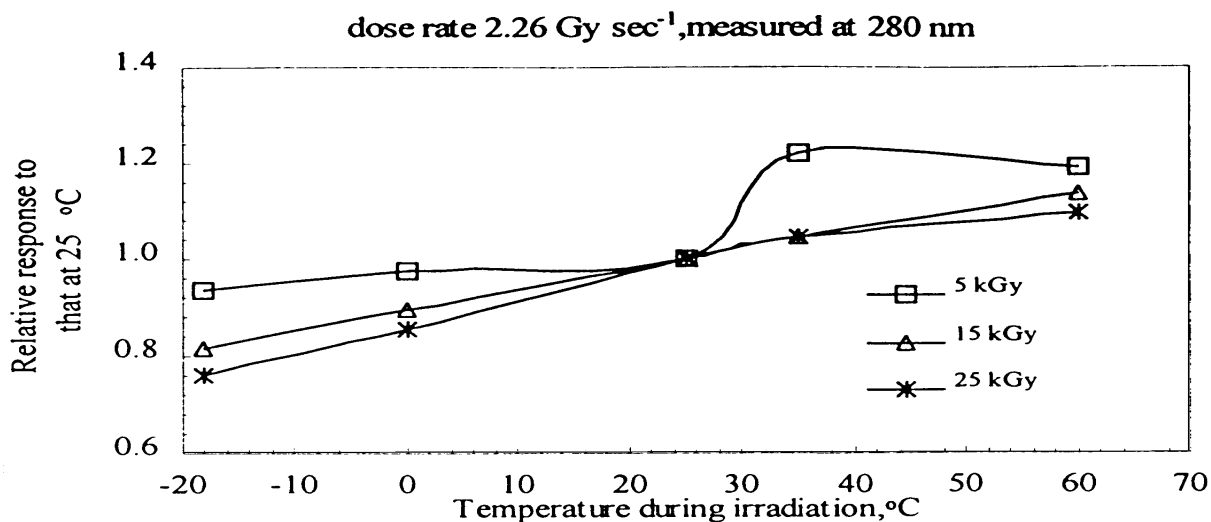


FIG. 9. Temperature dependence, relative to that at  $25^\circ\text{C}$ , of gamma response of CTA at dose rate  $2.26 \text{ Gy sec}^{-1}$  where the absorbance per unit thickness was measured at 280 nm.

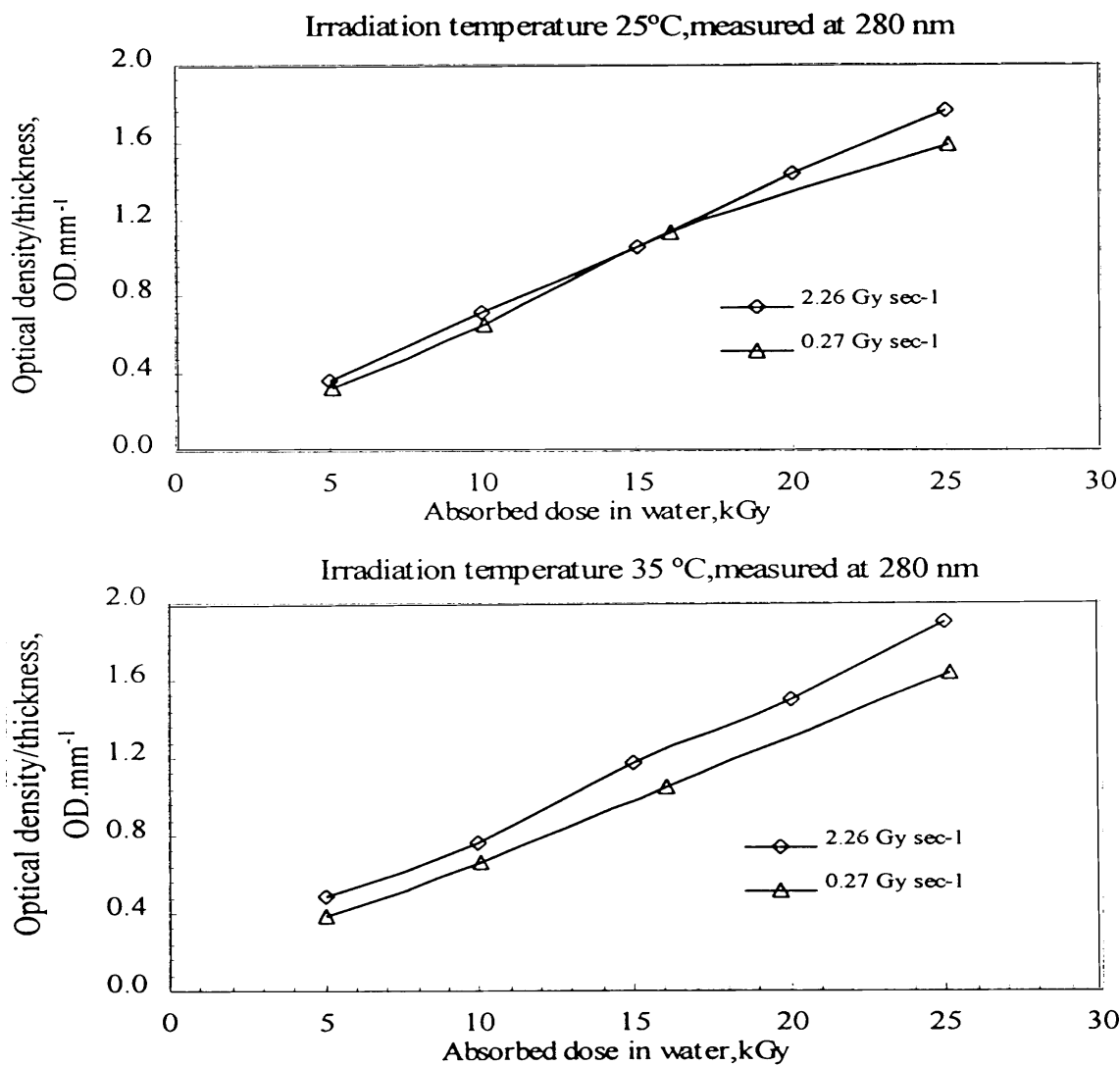


FIG. 10. Combined effect on the response of cellulose acetate (CTA) at dose rate of  $2.26$  and  $0.27 \text{ Gy sec}^{-1}$  with the irradiation temperature of  $25$  and  $35^\circ\text{C}$ .

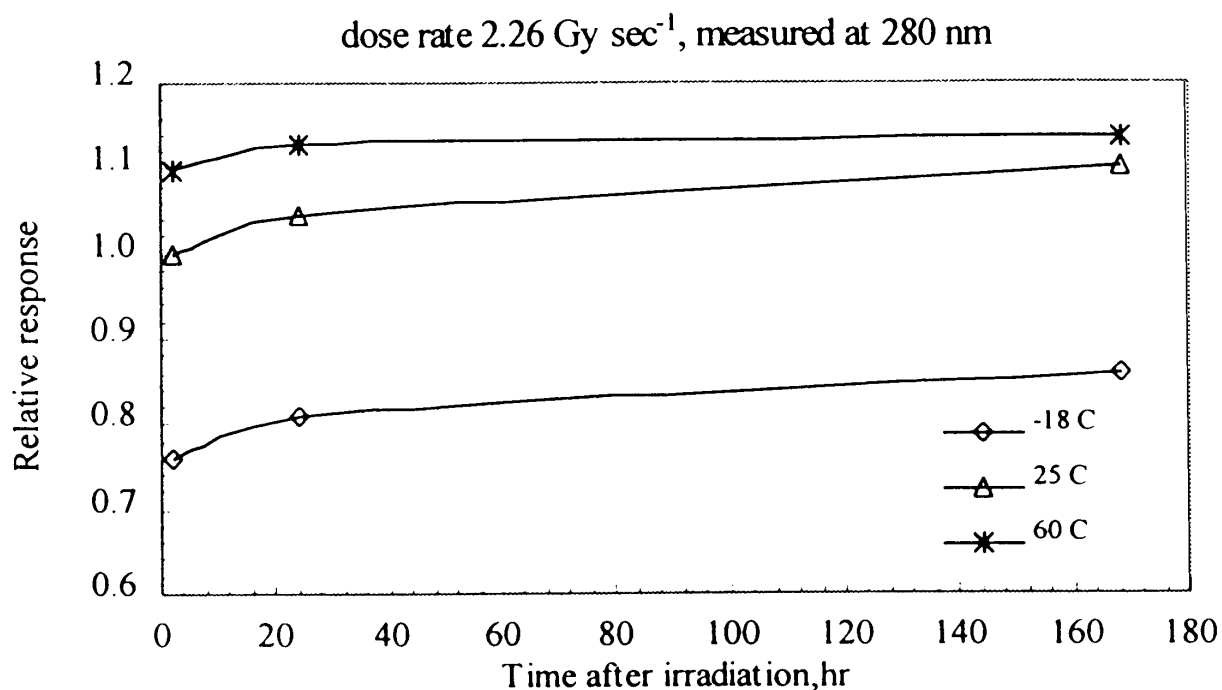


FIG. 11. Short term post-irradiation studies of cellulose acetate (CTA) at 25 kGy, analysis time 2, 24 hours and 7 days after irradiation for the dose rate  $2.26 \text{ Gy sec}^{-1}$ .

## 2.5. Radiochromic film FWT-60-00 dosimeter [14]

The transparent radiochromic film, FWT-60-00 batch 5E4 and 7F7, are commercially supplied in  $10 \text{ mm} \times 10 \text{ mm} \times 40\text{--}50 \text{ }\mu\text{m}$  by Far West Technology, Inc. They are thin colourless films that change to deep blue upon irradiation to nominal absorbed doses from 1 to 50 kGy.

### 2.5.1. Combined effects of irradiation temperature and dose rate and short term post-irradiation studies

Three films for each irradiation were held between two electron equilibrium layer of 3-mm thick acrylic and sealed at 60% r.h. in the polyethylene-aluminium foil laminate for protection against changing environmental condition specially humidity. The pre-irradiation absorbance for each dosimeter was read using Shimadzu UV-3101PC UV/vis spectrophotometer at 605 nm. The dosimeters were conditioned for 1 h at each irradiation temperature before irradiation and then irradiated using a double-walled glass dewar flask for temperature control. The irradiations were carried out using  $^{60}\text{Co}$  Gammabeam 650 panoramic source at dose rate of 4.04 Gy/s for the dose range from 5 up to 50 kGy at the irradiation temperatures of  $-18, 0, 35$  and  $50^\circ\text{C}$  using the coolants as shown in Table II. Fluctuation of the irradiation temperature was within  $\pm 3^\circ\text{C}$ . The irradiated dosimeters were kept at  $25^\circ\text{C}$  for 2 h before absorbance reading and reread again at 24 h and 7 d after irradiation for short term post-irradiation studies. For the combined effect studies, the irradiations were carried out using two  $^{60}\text{Co}$  sources, gammacell 220 at absorbed dose rate of 2.7 Gy/s and Gammabeam 650 panoramic source at 0.18 Gy/s for the absorbed dose of 5 kGy at the following temperatures:  $-78, -18, 0, 25$  and  $60^\circ\text{C}$  using the same conditions as previous experiment.

### 2.5.2. Results and conclusion

Figure 12 shows temperature dependence of the response of FWT-60-00 batch 7F7 at dose rate 4.04 Gy/s for dose range 5 to 50 kGy over the irradiation temperature of -18 up to 50°C. The results of the dose rate effect in Fig. 13 indicate that the bias on a dose interpretation based on calibration at a high dose rate would be underestimated by another irradiation to an unknown dose at a lower dose rate. The effect of post-irradiation on the response of dosimeter in Fig. 14 shows insignificant.

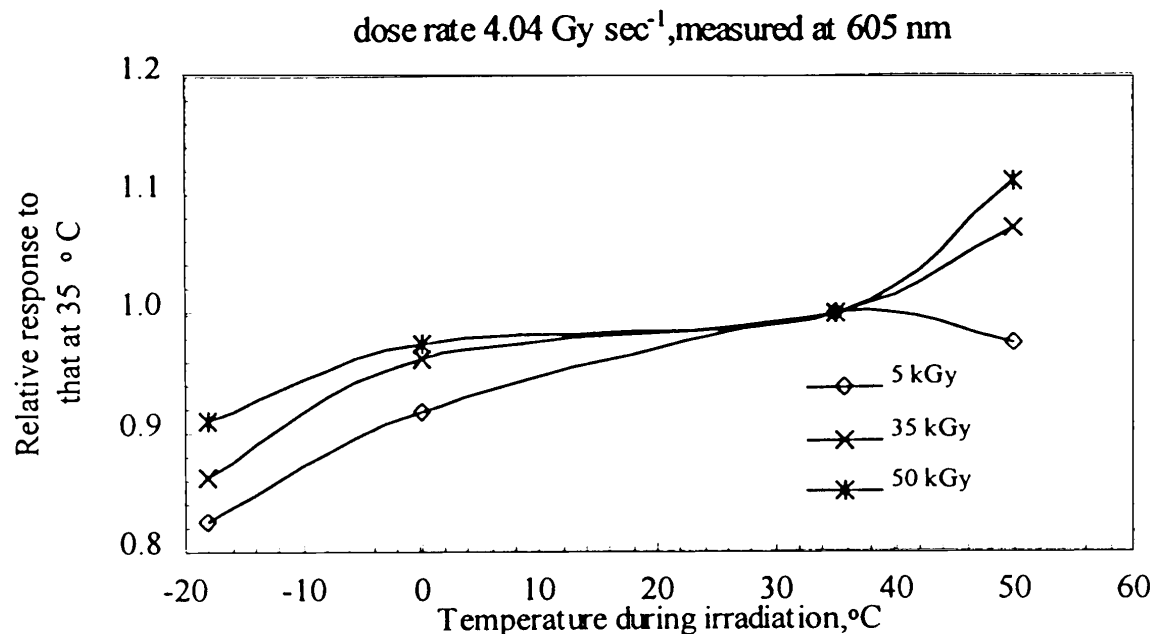


FIG. 12. Temperature dependence, relative to that at 35°C, of gamma response of radiochromic film FWT-60-00 batch 7F7 at dose rate 4.04 Gy sec<sup>-1</sup>. Where the absorbance per unit thickness was measured at 605 nm.

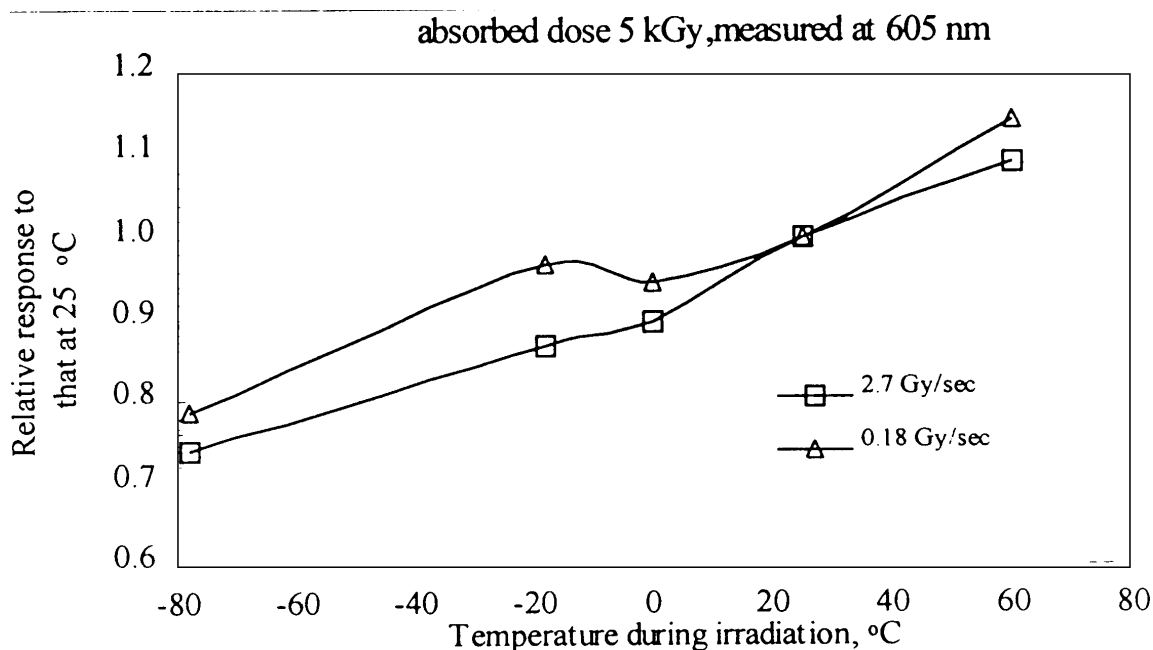


FIG. 13. Combined effect on the response of radiochromic film FWT-60-60 batch 5E4 (at 5 kGy) at dose rate 2.7 and 0.18 Gy sec<sup>-1</sup> with the irradiation temperature from -78 to 60°C.

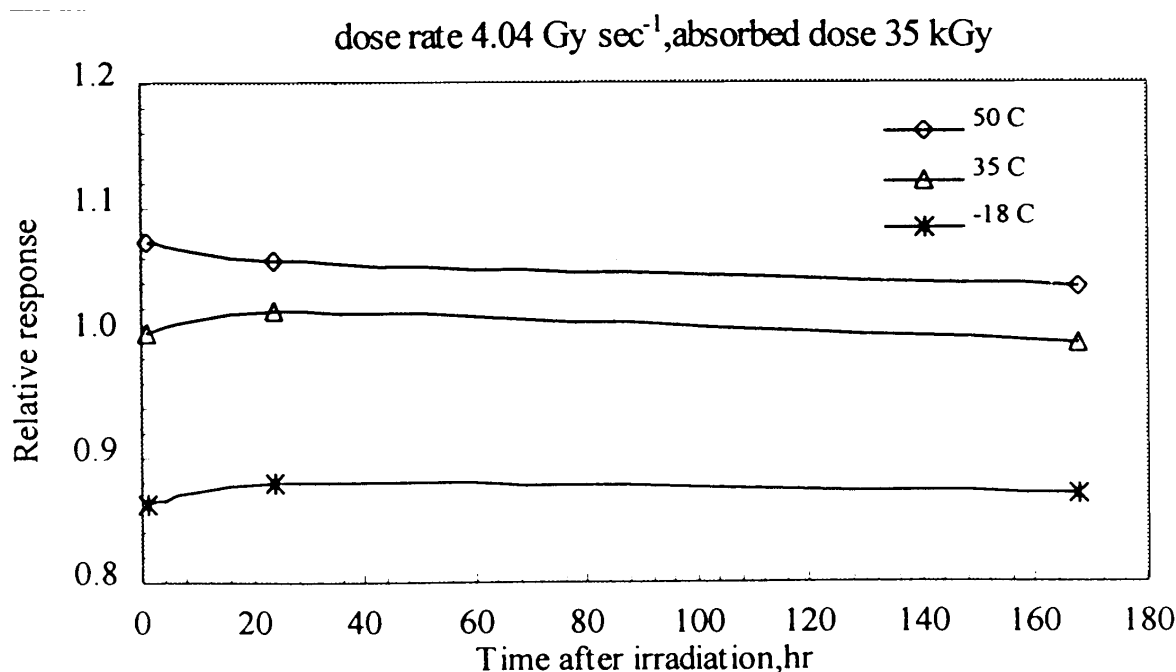


FIG. 14. Short term post-irradiation studies on gamma response of radiochromic film FWT-60-00 batch 7F7 at 35 kGy, analysis time 2.24 hours and 7 days after irradiation for the dose rate  $4.04 \text{ Gy sec}^{-1}$ .

## 2.6. Red Perspex 4034 polymethylmethacrylate dosimeter [15]

Harwell Red 4034 batch DA is a dyed polymethylmethacrylate dosimeters which are commercially available in the puncture-resistance polyester/aluminium/polyethylene laminated sachets for protection against changing environmental condition.

### 2.6.1. Combined effects of irradiation temperature and dose rate and short term post-irradiation studies

The irradiations were carried out using  $^{60}\text{Co}$  Gammabeam 650 panoramic source at absorbed dose rate of  $4.04 \text{ Gy/s}$ . The dosimeters were conditioned for 1 h and irradiated at each temperature using a double-walled glass dewar flask and stored at room temperature immediately after the end of irradiation. The dosimeters were irradiated for the dose range of 5 up to 50 kGy at the following temperatures:  $-18$ ,  $0$ ,  $35$  and  $50^\circ\text{C}$ . Fluctuation of the irradiation temperature was within  $\pm 3^\circ\text{C}$ . Table II shows the conditions in the dewar flask. The irradiated dosimeters were kept at about  $25^\circ\text{C}$  for 2 h before spectrophotometric (Shimadzu UV-3101PC) analysis and reread again at 24 h and 7 d after irradiation for short term post-irradiation studies. For the combined effect studies, the irradiations were carried out using two  $^{60}\text{Co}$  sources, gammacell 220 at absorbed dose rate of  $2.7 \text{ Gy/s}$  and Gammabeam 650 panoramic source at  $0.18 \text{ Gy/s}$  for the absorbed dose of 5 kGy at the following temperatures:  $-78$ ,  $-18$ ,  $0$ ,  $25$  and  $60^\circ\text{C}$  using the same conditions as previous experiment.

### 2.6.2. Results and conclusion

The results in Figs 15 and 17 show only slight variation in response of the PMMA Red 4034 on post-irradiation behaviour over the irradiation temperature of  $-18$  up to  $50^\circ\text{C}$  for the

period of 2 h to 7 day. The temperature dependence at 5 kGy shows similar behaviour for the two dose rates investigated, 0.18 and 2.7 Gy/s, between 0°C and 25°C, but outside of this temperature range the dose-rate dependence was found to be significant as shown in Fig. 16.

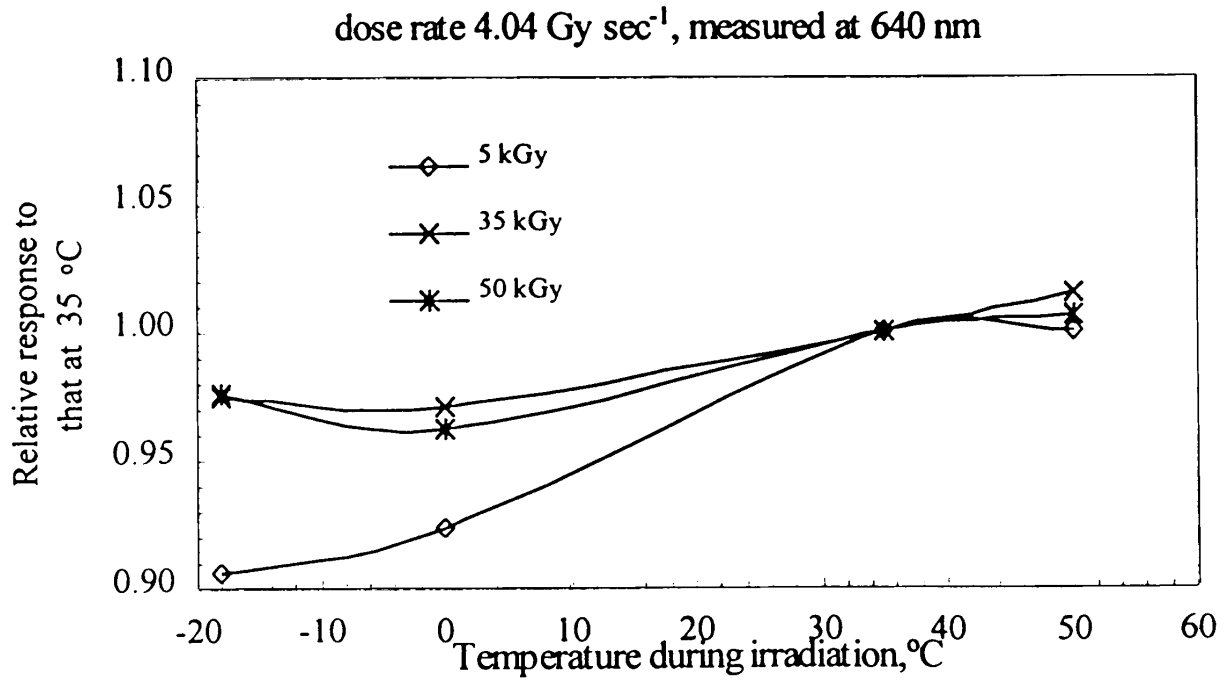


FIG. 15. Temperature dependence, relative to that at 35°C of gamma response of red perspex 4034 batch DA at dose rate 4.04 Gy sec<sup>-1</sup>. Where the absorbance per unit thickness was measured at 640 nm.

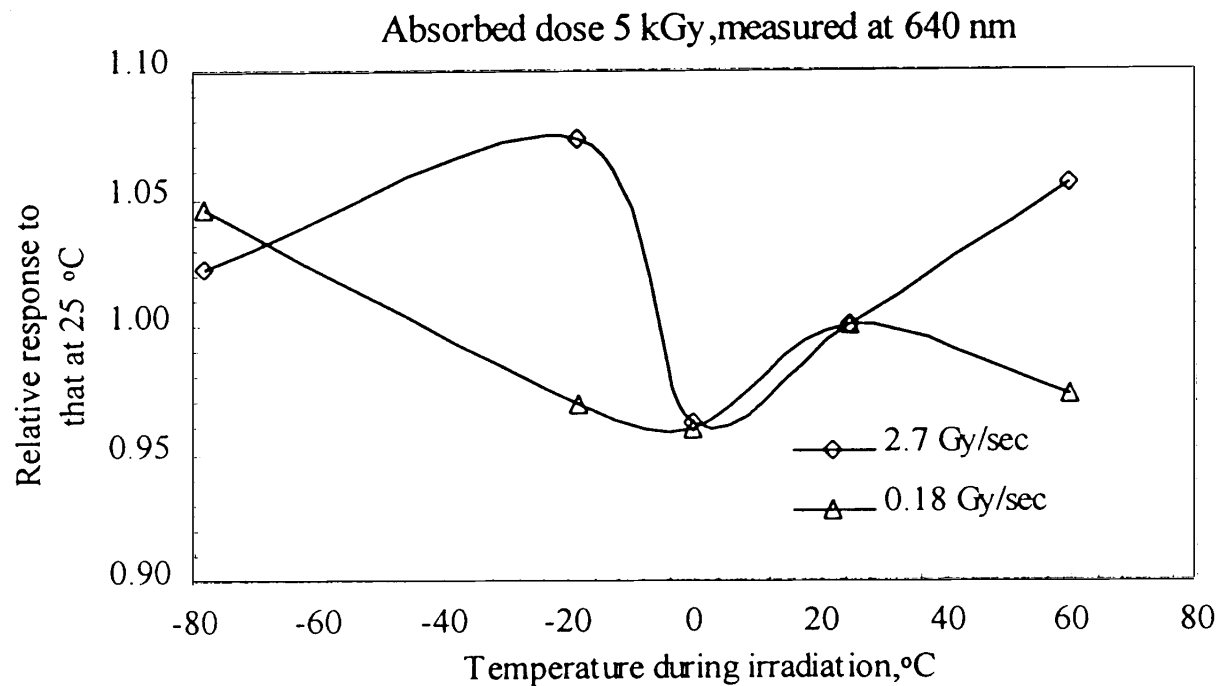


FIG. 16. Combined effect on the response of red perspex 4034 batch DA (at 5 kGy) at dose rate of 2.7 and 0.18 Gy sec<sup>-1</sup> with the irradiation temperature from -78 to 60°C.

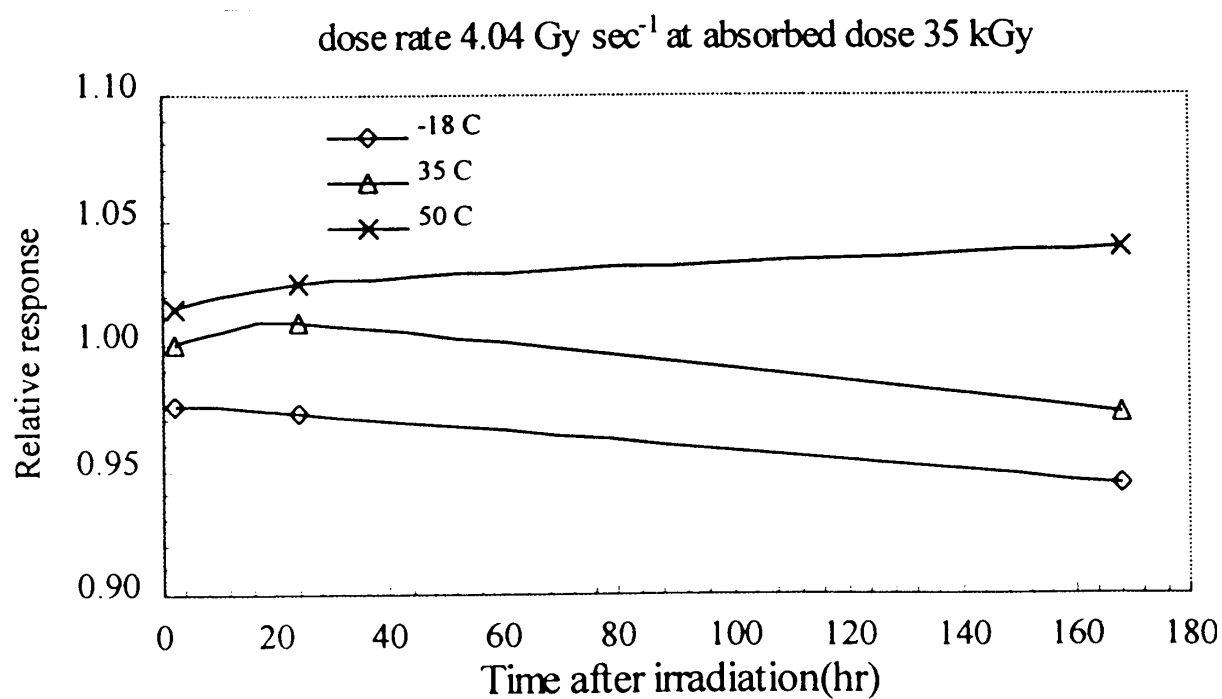


FIG. 17. Short term post-irradiation studies on gamma response of red perspex 4034 batch DA at 35 kGy, analysis time 2, 24 hours and 7 days after irradiation for the dose rate  $4.04 \text{ Gy sec}^{-1}$ .

### 3. CONCLUSION

As the national calibration laboratory for high-dose dosimetry, this research work has helped us to appreciate the influence of various parameters on the performance of many types of commercial routine dosimeters. Assuring that the calibration conditions are as close to the conditions of use as possible will increase the dosimetry accuracy and reliability for quality assurance in radiation processes.

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# REFERENCE DOSIMETRY AND CALIBRATIONS WITH X AND GAMMA RAYS AND ELECTRON BEAMS

W.L. McLAUGHLIN

Ionizing Radiation Division, Physics Laboratory,  
National Institute of Standards and Technology,  
Washington, D.C., United States of America

## Abstract

For quality control, validation, and research and development in radiation processing with photons and electrons, standardized dosimetry, calibration and traceability are essential. Difficulties in achieving the objectives of reproducible and accurate high-dose dosimetry include the following influences on the measurement uncertainties: geometrical and environmental variabilities, dose rate and energy dependence, dosimeter system instabilities, type of radiation, type of analysis, radiation source factors, and precision of dosimeter supplies. The objectives under this Research Agreement are: 1) to develop special calorimetric approaches as means of calibration and traceability; 2) to evaluate the individual and combined uncertainties of a key radiation processing dosimetry system used by NIST for x- and gamma rays and electron beams, namely, alanine analyzed by EPR spectrometry. The NIST alanine dosimeter is studied as a reference transfer dosimetry system in terms of temperature dependence in its response to  $^{60}\text{Co}$  gamma rays and the influence of relative humidity on post-irradiation stability.

## 1. INTRODUCTION

The research effort under this agreement with the IAEA has been aimed at establishing improved means of traceability of dosimetry at high doses applicable for a relatively wide range of doses needed for radiation processing. The research at NIST has focused on two main goals: 1) Establishment of a calorimetric means calibration of the response of the high-dose dosimeters in photon and electron radiation fields; 2) evaluating certain sources of measurement uncertainty for the dosimetry systems, such as environmental influences, and post-irradiation stability.

The calorimetry systems are of two types: 1) For ionizing photons (e.g. a "teletherapy" vertical beam gamma ray source), a water calorimeter is used [1]; 2) for electron beams ( e.g. a 10 MeV linear accelerator), a modular graphite calorimeter is used [2]. The NIST alanine dosimetry system consists of 90% (by mass) microcrystalline L- $\alpha$ -alanine (Aldrich) dispersed in a binder consisting of 10% (by mass) polyethylene (Polysciences, MW = 700, 60  $\mu\text{m}$ ) [3].

The alanine dosimeters are analyzed routinely by an ESC 106 EPR spectrometer (Bruker Instruments) with a 4103TM cavity at room temperature, the modulation frequency set at 50 kHz and the microwave power at 0.25 kW.

## 2. CALORIMETRY

The water calorimeter was previously developed by Domen as the national standard for absorbed dose measurement of gamma rays at doses  $>1$  Gy [1]. It is used to determine the absorbed dose rate in water, for the reference vertical beam at a source-to-detector distance of 100 cm at a depth of 5 cm in a water phantom. By using a scaling theorem [4], this gamma ray dose rate is converted to dose in water at a depth of 4.87 cm in a polystyrene phantom at which location dosimeters are calibrated under electron equilibrium conditions. The details of the procedures for achieving calibration traceability for ionizing photon irradiations by irradiating calibrated NIST alanine dosimeters in specified geometries of different gamma ray

irradiators (e.g. GammaCells) are given in a NIST Special Publication [5]. In an associated NIST Special Publication [6] are given tabulations of the uncertainties of absorbed dose measurements by different gamma ray irradiated dosimeters under this traceability procedure, an example of which is given in Table I.

TABLE I. UNCERTAINTIES IN ABSORBED DOSE (IN WATER) VALUES FOR ALANINE TRANSFER DOSIMETERS

Source of Uncertainty	Type A (%)	Type B (%)
Alanine single-hole cup dose rate	0.36	0.47
Alanine dose measurement	0.71	0.22
Irradiation temperature correction	----	0.10
Time between irradiation & analysis	----	0.25
Dose value from calibration curve	----	0.50
Combined separately	0.80	0.77
Combined relative standard uncertainty	.. 1.1	
Relative expanded uncertainty (k=2).....	.. 2.2	

TABLE II. MODULAR GRAPHITE CALORIMETER ESTIMATED MEASUREMENT UNCERTAINTIES

Component	Type A (%)	Type B (%)
Temperature Measurement:		
Calibration of quartz thermometer	----	0.05
Temperature readout reproducibility	0.03	----
Temperature readout noise	0.03	----
Thermistor resistance stability	----	0.05
Ohmmeter reproducibility and stability	----	0.05
Temperature calibration curve fit	----	0.1
Effects of mass of non-graphite components on temperature response	----	0.05
Specific Heat Capacity		
Measured graphite heat capacity	----	0.5
Specific heat data curve fit	----	0.1
Physical Properties of Graphite Disks		
Thickness measurements	----	0.3
Density measurements	----	0.2
Interpretation of Temperature Response		
Linear regression fit	----	0.07
Use of midpoint vs. equal area time of irradiation	----	0.08
Dose Conversion		
Graphite to water (stopping power ratio)	----	0.5
Estimated combined uncertainty ...	..0.81	
Expanded uncertainty (k=2) .....	..1.62	

A novel modular graphite calorimeter has been developed at NIST for calibration of reference and working standard dosimeters irradiated in high-energy electron beams (e.g. 10 MeV) [2]. It consists of a stacked array of eight high-purity graphite disk, each 1.8 mm thick and 50 mm in diameter, oriented such that the flat surfaces are perpendicular to the direction of the incident electron beam. As many as eight disks, each containing a dedicated calibrated thermistor thermally isolated from the adjacent ones by thin (0.5 mm) polystyrene rings, can be used in a rigid configuration. Once the individual graphite disk readings are established for a given electron beam geometry and spectrum, one of the interior rings at a penetration depth along the approximately linear rising portion of the electron depth-dose curve is substituted by a phantom graphite ring of the same dimensions. Using this arrangement, small dosimeters may be calibrated while held tightly in the central region of the phantom disk, using the corresponding dose ratios between the adjacent calorimeter modular rings upon beam irradiation. The details of the calibration procedures and traceability by this approach are given in reference [3]. The estimated uncertainties associated with electron beam absorbed dose measurements with this modular graphite calorimeter are given in Table II.

### 3. REFERENCE TRANSFER ALANINE DOSIMETRY SYSTEMS

Recent studies have been made on the influence of environmental effects, namely, temperature and relative humidity, on the response and post-irradiation stability of the NIST alanine/EPR dosimeters to gamma radiation [7, 8]. The overall results may be summarized as follows.

#### 3.1. Temperature effects [7]

Previous results in the literature have indicated that the irradiation temperature dependence of the gamma ray sensitivity of alanine dosimeters in terms of a *temperature coefficient factor* [defined as  $S(22C)/S(T)$  in units of %/K] is relatively well-behaved, that is, it is generally a constant positive value over a broad range of irradiation temperatures. However, the values of this factor reported in the literature can vary from about +0.1 to +0.4 %/K, depending mainly on the particular alanine dosimeter being used. For the NIST alanine pellet dosimeter, the temperature coefficient factor over an irradiation temperature range 10 to 50°C varies appreciably with the absorbed dose, as shown in Table III.

TABLE III. TEMPERATURE COEFFICIENT VS ABSORBED DOSE

Absorbed Dose (kGy)	Temp. Coefficient (%/K)
3	+0.159
10	+0.148
20	+0.132
40	+0.142
60	+0.161
80	+0.178
100	+0.194

### 3.2. Relative humidity effects on stability [8]

Although the radiation-induced EPR signal used for dosimetry is considered quite stable for at least six months after irradiation under normal laboratory storage conditions, recent results at NIST show certain departures at high doses and high relative humidities. Studies of the influence of relative humidity during post-irradiation storage show in general the following properties regarding stability of the EPR signal:

- The higher the relative humidity, the greater the fading and fading rate over storage periods up to one year.
- The higher the dose from 1 to 100 kGy, the greater the fading and fading rate.
- Short term instabilities up to several hours after irradiation: At relative humidities up to about 60%, the signal is stable to within approximately  $\pm 1\%$ ; at higher relative humidities, the signal increases during the first few hours while in the EPR cavity presumably because of drying of the pellet during this period.
- Long term instabilities up to one year after irradiation: A relatively steady rate of fading occurs after the first few days following irradiation. Table IV shows the degrees of fading of the EPR signal for a dose of 10 kGy at different relative humidities and storage times.

TABLE IV. FADING VS RELATIVE HUMIDITY

Relative Humidity (%)	Fading (%)		
	30 days	60 days	365 days
0	0.2	0.4	2.2
33	0.3	0.7	3.8
44	0.3	0.7	4.9
57	1.0	1.6	6.5
75	2.2	3.5	10.1
94	10.0	12.5	18.5

In summary, for the greatest degree of both short- and long term stability, the recommended relative humidity for storage as well as EPR readout of the irradiated EPR dosimeter pellets is about 33 %, which is typical of normal laboratory conditions. This is the case whether or not the dosimeter is analyzed in the presence of an adjacent ruby ( $\text{Cr}^{3+}$ ) reference standard sample [8].

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## PARTICIPANTS IN THE CO-ORDINATED RESEARCH PROJECT

- Biramontri, S.            Radiation Measurement Division,  
Office of Atomic Energy for Peace (OAEP),  
Bangkok, Thailand
- Chen, W.                Department of Chemistry, Radiation Research Centre,  
Beijing Normal University, Beijing, China
- Chu, R.                 MDS Nordion, Engineering and Technology,  
Kanata, Ontario, Canada
- Dolo, J.-M.            Commissariat à l'énergie atomique,  
DTA/DAMRI/LMRI, Gif-sur-Yvette, France
- Gupta, B.L.            Radiation Standards and Instrumentation Division,  
Bhabha Atomic Research Centre (BARC),  
Trombay, Mumbai, India
- Kojima, T.             Advanced Radiation Technology Centre,  
Japan Atomic Energy Research Institute (JAERI),  
Takasaki-shi, Japan
- Kovács, A.            Institute of Isotope and Surface Chemistry,  
Chemical Research Centre of the Hungarian Academy of Sciences,  
Budapest, Hungary
- McLaughlin, W.L.    Ionizing Radiation Division,  
National Institute of Standards and Technology (NIST),  
Gaithersburg, Maryland, United States of America
- Miller, A.             Risø National Laboratory,  
High Dose Reference Laboratory,  
Røskilde, Denmark
- Zagórski, Z.P.        Institute of Nuclear Chemistry and Technology,  
Department of Radiation Chemistry,  
Warsaw, Poland

**CONTRIBUTORS TO DRAFTING AND REVIEW OF THE  
FINAL REPORT OF THE CO-ORDINATED RESEARCH PROJECT**

Biramontri, S.	Office of Atomic Energy for Peace, Thailand
Chen, W.	Beijing Normal University, China
Chu, R.	MDS Nordion, Canada
Dolo, J.-M.	Commissariat à l'énergie atomique, France
Gupta, B.L.	Bhabha Atomic Research Centre, India
Kojima, T.	Japan Atomic Energy Research Institute, Japan
Kovács, A.	Institute of Isotope and Surface Chemistry, Hungary
McLaughlin, W.L.	National Institute of Standards and Technology, United States of America
Mehta, K.	International Atomic Energy Agency
Miller, A.	Risø National Laboratory, Denmark
Zagórski, Z.P.	Institute of Nuclear Chemistry and Technology, Poland-



