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Standardized methods to verify absorbed dose in irradiated food for insect control

Proceedings of a final Research Co-ordination Meeting organized by the Joint FAO/IAEA Division of Nuclear Techniques in Food and Agriculture and held in Cascais, Portugal, 30 March–3 April 1998

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FOREWORD

Irradiation to control insect infestation of food is increasingly accepted and applied, especially as a phytosanitary treatment of food as an alternative to fumigation. However, unlike other processes for insect control, irradiation does not always result in immediate insect death. Thus, it is conceivable that fresh and dried fruits and tree nuts, which have been correctly irradiated to meet insect disinfestation/quarantine requirements, may still contain live insects at the time of importation. There is, however, a movement by plant quarantine authorities away from inspecting to ensure the absence of live insects in imported consignments towards examining through administrative procedures that a treatment required by law has been given. Nevertheless, there is a need to provide plant quarantine inspectors with a reliable objective method to verify that a minimum absorbed dose of radiation was given to supplement administrative procedures. Such an objective method is expected to bolster the confidence of the inspectors in clearing the consignment without delay and to facilitate trade in irradiated commodities.

The Joint FAO/IAEA Division of Nuclear Techniques in Food and Agriculture initiated a co-ordinated research project (CRP) in 1994 to generate data on the verification of absorbed dose of irradiation in fresh, dried fruits and tree nuts for insect disinfestation/quarantine purposes. A standardized label dose indicator available commercially was used to verify the minimum/maximum absorbed dose of the irradiated commodities for these purposes as required by regulations in certain countries. It appears that such a label dose indicator with certain modifications could be made available to assist national authorities and the food industry to verify the absorbed dose of irradiation to facilitate trade in such irradiated commodities.

This TECDOC reports on the accomplishments of this co-ordinated research project and includes the papers presented by the participants of this CRP at the final Research Co-ordination Meeting held in Cascais, Portugal, 30 March–3 April 1998.

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EDITORIAL NOTE

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SUMMARY

1. INTRODUCTION

Global trade in food and agricultural commodities has become more liberal following the GATT Uruguay Round and subsequent establishment of the World Trade Organization (WTO) in 1995. Among the agreements attached to the agreement establishing the WTO, the Agreement on the Application of Sanitary and Phytosanitary Measures (SPS) is of particular relevance to international trade in irradiated food. The SPS Agreement aims at protecting the health and lives of humans, animals and plants based on measures established by sound science. The Agreement also recognizes standards, guidelines, and recommendations of international organizations including the Codex Alimentarius Commission, the International Plant Protection Convention, and the International Office of Epizootics, to assist the WTO in settling trade disputes. It is expected that the outcome of this research project will make a positive contribution to trade in food and agricultural commodities.

At the final Research Co-ordination Meeting participants deliberated on the global developments on the application of food irradiation with emphasis on its role as a method to control insect infestation of food. Radiation processing is increasingly being accepted and applied in many countries as a method to reduce food losses, control certain foodborne diseases, and facilitate wider food trade. As of 1998, 44 countries have approved the irradiation of a number of food products for consumption and over 30 countries are currently using the technology for several food products for commercial purposes.

Among several applications of food irradiation, its use as an alternative to the fumigation of food is gaining increasing recognition and utilization. Major importing countries have shown increasing concern about imported food and agricultural commodities which may harbour foreign pests. Consequently, strict measures have been introduced to prevent entry/establishment of such pests in their countries. In addition, there is increasing concern among consumers regarding pesticide residues in food. Fumigation of food and food ingredients for insect control by various chemicals, such as ethylene dibromide (EDB), methyl bromide (MB), and ethylene oxide (ETO), has either been prohibited or is being increasingly restricted in most advanced countries for health, environmental, or occupational safety reasons. EDB was banned by the US Environmental Protection Agency in 1984. The importation from other countries of any food treated with EDB for sale in the USA is also prohibited. Most countries have followed the USA in banning the use of EDB for the fumigation of food and food ingredients for the purpose of insect disinfestation.

MB is the most widely used fumigant for food and agricultural products against pests such as insects and nematodes and it will most likely suffer the same fate as EDB. MB has been listed under the Montreal Protocol (an international treaty for the regulation of ozone depleting substances worldwide, under the auspices of the United Nations Environmental Programme) as one of the substances which causes depletion of the ozone layer. At a meeting held in Montreal in 1997, the Parties to the Montreal Protocol agreed to the following phaseout schedules for MB:

Advanced countries:	25% reduction by the year 1999
	50% reduction by the year 2001
	70% reduction by the year 2003
	Phaseout by the year 2005 (with exemption for critical uses)
Developing countries:	20% reduction by the year 2005
	Phaseout by the year 2015

The use of MB for quarantine purposes and for preshipment fumigation was exempted from the phaseout schedule under the Montreal protocol.

The Clean Air Act of the USA promulgated in 1990 would have required MB to be deregistered by 31 December 2000 because of its ozone depleting potential. Indeed, the US Environmental Protection Agency published the final rule on 30 November 1993 to terminate the production and consumption of MB by this date. However, the US Congress decided in 1998 to phase out MB according to the same schedules agreed under the Montreal Protocol. Therefore, the production and consumption of MB in the USA will have to be abided by the phaseout schedules of the Montreal Protocol. The question remains, however, on the availability of MB for quarantine and preshipment purposes as well as its cost after this chemical is no longer produced in advanced countries starting 2005.

ETO, another widely used fumigant for decontaminating dry food ingredients, particularly spices and dried vegetable seasonings, was banned by the European Union starting from 1 January 1991. The US EPA issued a final rule which prohibited the use of ETO for fumigating processed spices on 22 March 1996. However, the EPA has not enforced the rule pending further review of data submitted by the spice industry. The use of irradiation to ensure hygienic quality of spices and dried vegetable seasonings to replace ETO fumigation has increased significantly in recent years.

Irradiation has been demonstrated as an effective replacement for the fumigants mentioned above. Low dose irradiation between 0.2 and 0.7 kGy can effectively control insect infestation of grain and other stored products and unlike fumigation, irradiation does not leave any residues in or on the products. To prevent reinfestation, grain and stored products must be properly packaged in insect proof containers.

Low dose irradiation is also being recognized as an alternative to EDB and MB fumigation of fresh agricultural products in order to overcome quarantine barriers in trade. A minimum dose of 0.15 kGy is effective as a quarantine treatment for fresh fruits and vegetables against fruit fly of the Tephritidae family, and a minimum dose of 0.3 kGy effective against other insect species. Unlike other competing technologies, irradiation is a broad spectrum quarantine treatment as it is not specific to insect species or host commodities. Its application as a quarantine treatment of fresh horticultural produce is endorsed by regional plant protection organizations which operate within the framework of the International Plant Protection Organization, i.e. North American Plant Protection Organization (NAPPO), European and Mediterranean Plant Protection Organization (EPPO), Asian and the Pacific Plant Protection Commission (APPPC), etc. The US Department of Agriculture (USDA) issued a Notice of Policy on 15 May 1996 which permits irradiation as a quarantine treatment of fresh fruits and vegetables against fruit flies regardless of host commodities. In addition, the USDA issued a specific regulation in 1997 permitting the use of irradiation for papaya, carambola and lychee against fruit flies from Hawaii. Countries which are members of the Association of Southeast Asian Nations (ASEAN) are also adopting a unified protocol on irradiation as a quarantine treatment for fresh horticultural produce.

Small scale commercial irradiation and sale of irradiated fresh fruits from Hawaii, with special permission from the USDA, has been carried out successfully in the USA since 1995. By the end of 1999, a total of some 350 metric tonnes of such fruits have been sold in many retail outlets, especially in the midwest of the USA, apparently without consumer opposition.

2. NEEDS TO VERIFY ABSORBED DOSE

National authorities, especially those responsible for plant protection and quarantine, specify in their regulations certain minimum doses of irradiation which need to be delivered to any part of the food in order to meet quarantine requirements. Unlike other processes for insect control, especially fumigation which is still widely applied, **irradiation of food for insect control in the dose range of 0.1 to 1.0 kGy does not always result in immediate insect death.** Thus, it is conceivable that fresh and dried fruits and tree nuts, which have been irradiated to meet insect disinfestation/quarantine requirements, may contain live insects at the time of importation. Normally, plant quarantine inspectors are instructed to reject any shipments which contain live insects. There is, however, a movement within the USDA to request their inspectors to examine only the certificate of consignment verifying that a required minimum radiation dose has been delivered to the food. Nevertheless, the availability of an objective method to assist plant quarantine inspectors in assuring that the minimum dose required by law has been achieved, would not only provide supplementary information to the administrative procedure but would also bolster the confidence of the inspectors in clearing the consignment without undue delay.

In commercial practice, fresh and dried fruits and tree nuts are normally packaged and transported in standardized containers/pallets. Given a specific bulk density of such food, it is possible to measure the minimum/maximum absorbed dose of the product in a standardized container. A standardized 'label dose indicator' could therefore be developed and affixed to the outside of the container and "read" by food/plant quarantine inspectors to verify the minimum absorbed dose. In instances where the maximum dose is regulated, it would be desirable if a label dose indicator could also verify the absorbed dose. For such a dose indicator to be workable, the radiation dose "read" from the label **must** be related to the minimum or maximum absorbed dose which has been previously determined through dosimetric validation processes.

According to the Codex General Standard for Irradiated Foods, any facility processing food by ionizing radiation must be licensed and registered. For the purpose of verification of absorbed dose from some reference dose readings, additional information on dose distributions achieved under the practices of the given facility and on the relationship of the dose measured at the chosen reference position to the critical minimum dose, needs to be established and related to the entries in the registry. Also, the statistical nature of the data has not yet been analyzed for the purpose of deriving confidence limits/error probabilities for decisions taken.

3. OVERALL OBJECTIVES

The co-ordinated research project (CRP) put particular emphasis on developing methods to verify the absorbed dose of irradiated fresh and dried fruits and tree nuts in trade. The overall objectives of the research were as follows:

- Validate dose distribution in containers used by trade under commercial conditions. In particular, to establish a link between statistical parameters of such a distribution and dose measurement at the reference position;
- Develop a quantitative label dose indicator and handheld or otherwise simple on the spot reader; and
- Standardize the application of such a dose indicator to an extent that the dose read can be accurately and reliably related to the minimum dose received in the container.

4. RESULTS OBTAINED

According to the objectives mentioned above, the following tasks were carried out under the scope of the CRP:

4.1. Validation

(a) Objective

To validate the application of label dose indicators to verify the absorbed dose in a batch or consignment of food. In particular, to establish a functional correlation between the value 'indicated' on the label and the characteristic parameters of the dose distribution throughout the goods.

(b) Criteria

The criteria included statistical parameters such as type of distribution, accuracy of the extrapolation to the extreme values, error probabilities for the estimated parameters, and finally the derivation of a decision function. The decision function would result in a 'yes/no' for the acceptance of an irradiated consignment and take into account the required confidence level for such decisions. It was understood that prerequisites must be met in order to render visual/subjective judgements meaningful for the purpose of verifying absorbed dose.

(c) Achievements

The main parameter to be observed is the 'source product geometry'. This includes the physical nature of the source and of the emitted radiation, as well as the arrangement of packages on pallets and on the transport system relative to the source and the direction of the radiation.

The dose distribution of the product is usually determined by using standardized packaging or containers normally employed in commercial practices for handling fresh and dried fruit and tree nuts. Such distributions vary by commodity, product and bulk density, individual packaging or bulk handling, and pretreatment for specific purposes. The physical extension of the products with reference to the dimensions of the source or the beam width is also to be considered.

Source geometry is related to the arrangement of the radioactive source elements in a source rack, plaque or cylinder. The physical dimensions of the source or the beam width with reference to the extension of the goods also need to be considered. The type of transport system (single and multiple pass; single, double, or multiple sided) is an important characteristic of the source product geometry as it significantly influences the position where the minimum absorbed dose is expected to occur.

In their contributions, the participants could only characterize a limited number of source product geometries. Pilot plants were mostly utilized; however, several studies included commercial scale industrial design facilities representing the geometries of:

- product overlap and plaque source arrangement;
- source overlap and cylindrical source arrangement;

- scanned unidirectional beams of electron and bremsstrahlung;
- single, double and multiple sided irradiation; and
- single and multipass irradiation.

Standardized commercial practices in handling and packaging fresh and dried fruit and tree nuts in countries of several participants were simulated in most cases. Depending upon availability, product geometries already prevailing in international trade were also utilized; examples are fresh fruit on cardboard trays stacked on pallets, or nuts in retail pouches assembled in cardboard boxes. The resulting dose distributions did not exhibit prominent deviations from normal (Gaussian) form and could be well characterized by the respective parameters of the Gaussian function. The number of studies presented was insufficient to relate the measurement of dose at a reference position together with its statistical parameters to the extrapolated extreme values (i.e. the minimum dose in particular) in a reliable manner.

A ‘label dose indicator’ is considered to transfer information by the appearance of the label which is equivalent to the signature of an inspector certifying his presence during the radiation processing. The certificate is issued on the correct execution of a predetermined and established procedure. Hence, a label and a certificate should convey the same information.

There are many possible product source geometries under different circumstances in commercial practices for fresh and dried fruit and tree nuts. Consequently, as many functional relationships as possible between dose at reference position and minimum absorbed dose in the goods need to be established. It is necessary that this collection of functional relationships is linked to the licensed and registered facility. If label dosimeters are used, their minimal value must refer to the related minimum value, and the built in decision function must reflect the source product geometry and the dose relationship. Hence, many types of label dosimeters with a range of transition points for the decision function need to be prepared; such sets of labels also need to be prepared for all values of required minimum doses.

An alternative to such dedicated label dosimeters could be ordinary dosimeters which can be read in situ and which are attached to standardized positions in/on the goods/products. Once the sets of functional relationships discussed above are established the appropriate decision functions could be built into a dedicated reader suitable for all situations/geometries. Most suitable for such purposes would be any single type of dosimeter with a wide dynamic range of dose values. Some such materials for in situ use are now becoming available commercially while others have been prepared on laboratory scale by some participants of this CRP. However, the mathematical deduction of the decision function and the validation under circumstances of commercial application have yet to be achieved.

4.2. Evaluation of label dose indicators and dosimetry systems

(a) Objectives

To evaluate existing label dose indicators suitable for verifying absorbed dose of irradiated fresh and dried fruits and tree nuts in trade. In addition, to investigate the application of other promising dose indicator systems for the same purpose.

(b) Criteria

Indicator labels must be proven suitable for visual determination that adequate irradiation treatment of fresh and dried fruits and tree nuts was applied for quarantine purposes. In order for this to be evaluated, abrupt change of the optical properties at the declared threshold dose is indispensable. On the other hand, the basic criterion expected of a successful dosimetry system, to be applied for the same purpose, by any objective readout method is the operability of the chosen dosimetry system in the dose range of interest.

(c) Achievements

ISP STERIN-125 and STERIN-300 irradiation label dose indicators were studied by both visual evaluation and instrumental methods such as spectrophotometry in transmitted and reflected light modes. It was shown that visual examination was able to give an indication of irradiation at the nominal dose ±30%. Spectrophotometry in the absorbed light mode showed that saturation at the maximum absorption occurred at a dose approximately 20% lower than the declared threshold values. Similar results were found in the measurements of traversed distances in chromaticity space, as well as in the measurements of brightness in terms of the CIE (Commission Internationale d´Eclairage) dimensions.

The usefulness of some other dosimeters as indicators of irradiation treatment for quarantine purposes was also evaluated. The dose response of the yellow PMMA dosimeters in the dose range 50–300 Gy, as measured by spectrophotometry, was adequate for that purpose, while the clear PMMA dosimeters were not satisfactory. The radiation induced transient electric current across the pn junction of bipolar power transistor in plastic packaging could also be used in the dose range of interest. However, neither PMMA nor the semiconductor type dosimeters were suitable for visual evaluation in the form of labels.

Several new dosimetry systems (Sunnar films, radiochromic dye in a solid matrix) based on fluorimetric and photometric readout were developed. While adequate dosimetric properties of the newly developed systems were established, their usefulness for the visual evaluation currently remains of limited value.

4.3. Development of readers

(a) Objectives

To develop a simple hand-held dosimeter reader capable of measuring absorbed dose in the range of interest from a dosimetric label/system placed on the outer surface of a standard product box/unit in a well defined reference position. This type of reader could provide an objective judgement using several analytical evaluation methods, since the interpretation of existing label dose indicators appears to be ambiguous and biased.

(b) Criteria

The reader must be able to give reliable dose readings in the range of 0.1 to 1.0 kGy with the possibility of being interfaced with a PC for data processing and documentation. The performance of such a reader should be controlled by using appropriate standards dedicated to the type of measurement.

(c) Achievements

The methods studied by the participants of the CRP for the design of the reader involve colorimetric, fluorimetric, oscillometric, and DC current measurement.

— The radiation induced colour change of label dose indicators (ISP STERIN) placed on the outer surface of product units can be evaluated by colorimetric analysis. A simple prototype hand-held reflectometer (consisting of two parts, i.e. the handset and the display unit) operating in the visible range has been built and is currently being tested. Another reflectometric system has also been designed and tested. The latter is under further development with the aim of converting it to a miniaturized hand-held label dose indicator reader for routine application.

— Commercial bipolar transistors have been selected and tested at laboratory level for the measurement of absorbed dose in the range of 50 Gy to 5 kGy. In order to retrieve the data, a portable DC current measuring instruments has been constructed and tested. Further development of this measuring instrument will allow the measurement of the transistors even on the outer side of the product unit.

— A digital, programmable, portable fluorimeter has also been designed, built and tested for measurement of the recently developed "Sunna" dosimeters based on the measurement of optically stimulated luminescence. These dosimeters have the potential of measuring doses in the range of 5 Gy to 100 kGy by inserting them into the slot of the portable fluorimeter. The system for measuring the irradiated dosimeters on the outer side of the product units is under development with a view to constructing a hand-held measuring device.

— Other dosimeter systems, like thermoluminescence (TL), can also be used to verify the absorbed dose of fresh and dried fruit and tree nuts, but without the potential application as label systems.

5. RECOMMENDATIONS

1. Efforts should be made to collect additional relevant information on the variability and the statistical properties of dose distributions for various geometries which occur under commercial practices for fresh and dried fruit, and tree nuts and in individual irradiation facilities. The information collected should be analyzed so that accurate and reliable conclusions may be provided to users/inspectors.

2. The development of promising new dosimetry/indicator systems should be encouraged. The existing STERIN label dose indicators should be developed with a steeper gradient of the transition at the nominal dose. For the indicator labels to be of practical use for most industrial irradiation facilities there is a need to develop dose indicators (for a wide range of nominal dose values) which are more flexible than the prototypes currently available. These could use the principle of ordinary and qualified dose measurement and current metrological practices.

3. There is a need to develop dedicated readers which incorporate all the established relationships from source product geometries and the respective functionality between dose at a reference position and the minimum dose required for the intended purpose.

4. Development of hand-held dosimeter readers has proven to be of immense importance for the reading of absorbed dose by any type of dosimeter that has been tested and considered to be suitable for the work carried out under this CRP for the following reasons:

- The reflectometric instruments that have been presented and tested are already on the way to being miniaturized and transformed into hand-held readers. The completion of such a development is important for the evaluation of colour changing dosimetry labels which are already available as well as those under development.
- The resistive load switching equipment for DC measurements from the bipolar transistor has to be improved by miniaturizing and interfacing it with a PC. It is recommended that it be equipped with a hand-held probe that can retrieve dose information by simple touch of the transistor's connectors.
- It is also recommended that the recently developed fluorimetric system should be equipped with a reader capable of performing measurements on dosimeters attached to the outer side of the product boxes and interfaced with a PC for data processing.

ESTABLISHMENT OF TECHNOLOGICAL PARAMETERS FOR DISINFESTATION OF DRIED FRUITS

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Abstract. A study to determine the irradiation parameters for disinfestation of dried fruits: figs, pine nuts, raisins and walnuts has been carried out in the UTR cobalt-60 facility. The dose distribution in the UTR boxes was measured. Low doses for disinfestation (150 Gy–300 Gy) were studied, for the commercial practice simulation/validation higher doses were used (15 kGy–20 kGy). The absorbed dose uniformity ratio (U = Dmax/Dmin) determined was 1.16 up to 1.33 for the dried fruits studied. Different dosimetric systems were tested. Low dose range dosimeters: reference standard Fricke dosimeter, routine dosimeters: Harwell YR Gammachrome and China PMMAYL dosimeter. High dose range dosimeters: routine dosimeters: Harwell Amber Perspex and Clear Perspex purchased at the local market. Label STERIN indicators of 125 Gy and of 300 Gy were assayed to establish a simple and direct process for verification, by customs inspectors, of a prior irradiation treatment. These indicators change their visual message if the threshold dose has been delivered. The performance of STERIN 125 and STERIN 300 suggested that these label indicators could properly be used for doses of 125 Gy and 300 Gy respectively, or higher than these ones.

1. INTRODUCTION

Dried fruits are often infested with insects responsible for the deterioration of these fruits. Therefore, its commercial distribution are subject to quarantine periods in order to avoid potential dissemination of insect pests. Until some years ago fumigation was used to prevent or destroy insect pests on foodstuffs, but nowadays chemical treatments have started to be questioned, banned or restricted due to serious limitations of the technique and to the harmful residues for human health left in food.

Irradiation seems to be an effective alternative to chemical treatments. Irradiation eliminates or controls the insect infestation of dried fruits, avoiding quarantine periods and does not present undesirable residues [1, 2].

The aim of this work was to study the dose distribution of dried fruits: figs, pine nuts raisins and walnuts treated by gamma radiation [3]. For dose range disinfestation the fruits were irradiated in fixed position at a defined place in the cobalt-60 facility. To simulate/validate the commercial practice conditions the irradiation was carried out in the current production process of the UTR facility using an absorbed dose of 15 kGy up to 20 kGy.

In the low dose range (150 Gy–300 Gy) the dosimetric systems Fricke, Harwell YR Gammachromme and China PMMAYL were employed; the STERIN label indicators were tested. For commercial simulation/validation irradiation the dosimetric systems: Harwell Amber Perspex and Clear Perspex were used.

2. MATERIALS AND METHODS

2.1. Dried fruits: figs, pine nuts, raisins and walnuts

All dried fruits were purchased at the local market. They were packaged prior to the irradiation in small bags of "VASCOLANPA/PESUPER"[1] film. This five layers laminate is composed of polyamide and polyethylene and constitutes a barrier to the outside atmosphere.

Inside the bags 250 g or 500 g of figs, 300 g of raisins and 200 g of pine nuts and walnuts. Were packed, the same volumes as used in trade. The bags were oriented in parallel to the source and ranged in small cardboard boxes (18×18×18) cm^3. These boxes were subunits of the UTR standard boxes (40×40×40) cm^3. Each UTR box had room for eight subunits.

2.2. Irradiation

The irradiation was carried out in the cobalt-60 facility UTR [4–6]: a) at fixed position for disinfestation dose range (2.2.1); b) during processing for simulation of commercial practice (2.2.2).

2.2.1. Irradiation in fixed position

The irradiation in fixed position was performed in front of the irradiator (cobalt-60) at a distance of 1340 mm to the source, close to the radiationshielding wall in a wood shelf. This wood shelf corresponds to the lower part of a half UTR carrier and can accommodate two UTR boxes (40×40×80) cm^3 (Fig. 1).

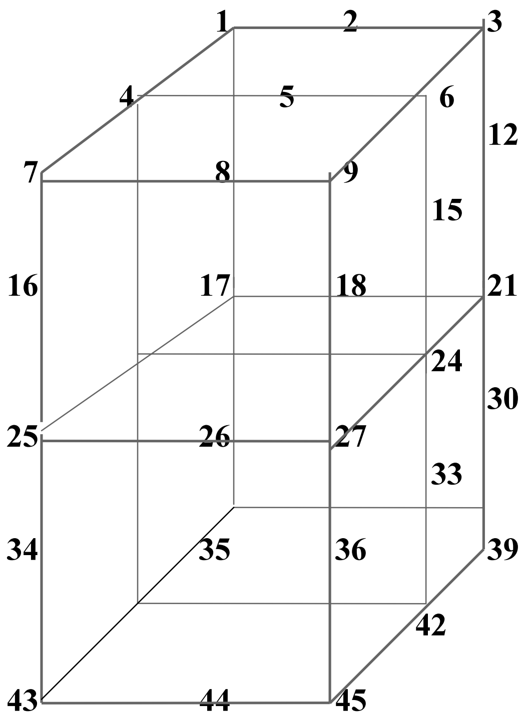

FIG. 1 Pair of UTR standard boxes moving together. Corresponding to a half UTR carrier and used as model for the studies at fixed position and for simulation of commercial practices.

[1]Obtained from VAESSENSCHOEMAKER, Portugal.

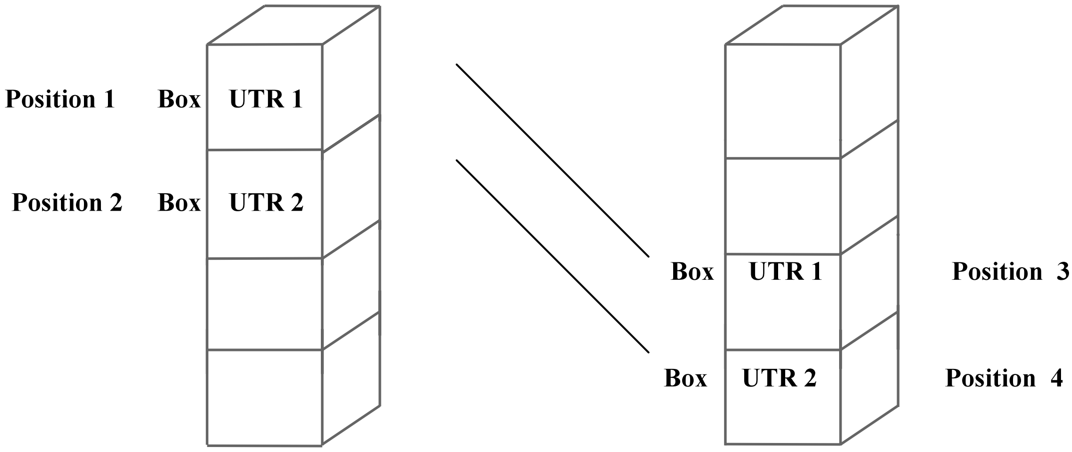

FIG. 2. Schematic representation of a UTR carrier holding four boxes. Movement of the boxes: position 1 and 2 during first cycle; position 3 and 4 on the second cycle.

A minimum absorbed dose of 140 Gy (>125 Gy) was used taking into consideration the radiation sensitivity of insects. The irradiation took 30 minutes, to improve the dose distribution, at half time a 180º rotation was done. The average dose rate was 0.35 $kGy.h^1$ for the dried fruits studied (2.1).

2.2.2. Irradiation in commercial practice

Two UTR boxes containing the dried fruits and the dosimeters were put in the upper position of a UTR carrier; after going through the plant they were moved to the lower part of the carrier and passed through the plant again in the second cycle (Figure 2).

2.3. Dosimeters

2.3.1. Low dose range

2.3.1.1. Ferrousammonium sulphate dosimeter (Fricke)

Fricke solution was prepared and calibrated following ASTM Standard E102692 [7]. The solution was irradiated in a geometry well defined (710 mm straight distance to the source; 1105 mm high to the soil). Three glass ampoules containing 5 ml were irradiated simultaneously during 2, 3, 4, 5, 6 and 7 minutes. The optical absorbance was red at 305 nm in a UV/VIS Spectrophotometer, at a temperature of 25ºC.

The data obtained were in agreement with the reference value (4.2 $kGy.h^1$/Dec. 92) for the same position measured by using an ionising chamber calibrated to a primary standard.

2.3.1.2. YR Gammachrome dosimeter

The Harwell YR Gammachrome dosimeter (Batch 3/sealed) was irradiated at the same geometry described in 2.3.1.1. The calibration curve built covered the dose range 0.1 kGy up to 3 kGy. The irradiation followed the supplier recommendations. This dosimeter was used at the UTR facility as a routine dosimeter for low absorbed doses applications.

2.3.1.3. PMMAYL dosimeter

The PMMAYL dosimeter was supplied by the Institute of Application of Atomic Energy, China where it is developed (dose range 0.05 kGy–1.00 kGy). The PMMAYL dosimeters (10×40 mm^2; 13 mm thicness) were sealed in aluminum bags and calibrated at the same geometry used in 2.3.1.1. The calibration curve built for this study covered dose range 0.05 kGy up to 0.40 kGy. It is a routine dosimeter for low absorbed dose applications, which has physical and chemical properties close to Harwell YR Gammachrome. The wavelength 530 nm recommended by that Institute was used. Specific optical absorbance was plotted against absorbed dose.

2.3.2. High dose dosimeters

2.3.2.1. Amber Perspex

The Harwell Amber Perspex type 3042 (Batch K/sealed), recalibrated at the same geometry used in 2.3.1.1, and in current use at the UTR facility for dose range 1 kGy up to 30 kGy was utilized. The wavelength 530 nm indicated by the supplier was used. Specific optical absorbance was plotted *versus* absorbed dose.

2.3.1.2. Clear Perspex

Clear Perspex was purchased at the local market and prepared at the UTR facility. A plate of Perspex were cut into small pieces (70×15 mm^2), which were dipped in a detergent solution, washed with a weak solution of acetic acid, rinsed well in tap water and in triple distilled water, then dried at 25ºC, put in aluminum bags and sealed [8]. The average thickness was 3.2 mm. The calibration curve was built in the dose range of 10 kGy up to 30 kGy. The irradiation was carried out at the same geometry used in 2.3.1.1. The wavelength selected was 290 nm and the reading temperature 25ºC.

2.3.3. Label indicators

STERIN 125 and STERIN 300 supplied by International Specialty Products (ISP)[1] were used. STERIN indicators are threshold indicators. A visual message “not irradiated” changes to “irradiated” if the threshold dose 125 Gy or 300 Gy is delivered or exceeded.

3. RESULTS AND DISCUSSION

3.1. Apparent density of dried fruits

In Table 1 are presented the apparent densities of subunits boxes and of UTR boxes for the different fruits.

[1] ISP: International Specialty Products, New York, USA.

TABLE. 1. APPARENT DENSITY OF DRIED FRUITS STUDIED

Dried Fruit	Apparent Density	
	SubUnit Box	UTR Std Box
Figs	0.40 g/cm^3	0.34 g/cm^3
Pinenuts	0.45 g/cm^3	0.33 g/cm^3
Raisins	0.33 g/cm^3	0.32 g/cm^3
Walnuts	0.31g/cm^3	0.23 g/cm^3

3.2. Irradiation in fixed position

3.2.1. Dose distribution

The absorbed dose measured by Fricke, Harwell YR Gammachrome and China PMMAYL dosimeters at different points/positions of the UTR boxes (Figure 1) containing dried figs are presented in Figure 3. The deviation between the dosimeters Fricke and the YR Gammachrome dosimeters are in agreement with the accuracy of the dosimeters [9]. The deviation of PMMAYL dosimeter is even more evidenced but seems acceptable for routine purposes as it is a in-house made dosimeter.

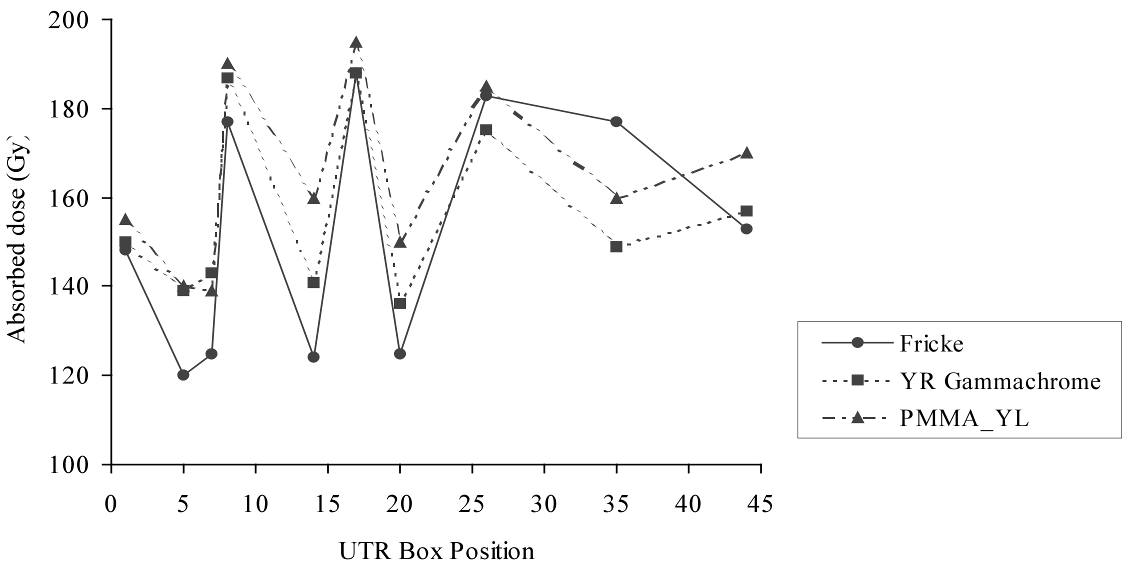

FIG. 3. Dose distribution of dried figs measured by different low dose dosimeters: Fricke (●); YR Gammachrome (■); PMMAYL (▲).

In Table 2 is presented the dose distribution in the UTR boxes, containing dried figs irradiated as described in (2.2.1) at fixed position. Minimum and maximum absorbed dose was measured by Fricke and Harwell YR Gammachrome dosimeters. The average dose uniformity obtained for both dosimeters does not show significant differences.

TABLE 2. DRIED FIGS DOSE DISTRIBUTION MEASURED WITH FRICKE AND YR GAMMACHROME DOSIMETERS

Dried Fruits	Absorbed Dose (Gy)					
Figs	Fricke Dosimeter			YR Gammachrome		
UTR	Dmin	Dmax	Dmax/ Dmin	Dmin	Dmax	Dmax/ Dmin
Box 1	160	230	1.49	130	170	1.33
Box 2	135	210		140	190	

In Table 3 is presented the different absorbed dose uniformity for the dried fruits studied measured with Harwell YR Gammachrome dosimeter. The irradiation was carried out at fixed position as described in (2.2.1), during 30 minutes. The average absorbed dose rate was $0.35\ kGy.h^{1}$.

TABLE. 3. ABSORBED DOSE UNIFORMITY FOR DIFFERENT DRIED FRUITS

Dried Fruits	YR Gammachrome Dosimeter Average Dose Uniformity
Figs	1.33
Raisins	1.23
Pine Nuts	1.18
Walnuts	1.20

3.2.2. Test of label indicators

The label indicators STERIN 125 and STERIN 300 were studied [10, 11]. It was possible to see the alteration in both label indicators STERIN 125 and STERIN 300. The word “NOT” disappeared and the word “IRRADIATION” was clear evidenced, when the absorbed dose delivered was respectively 100 Gy and 300 Gy.

TABLE.4. “STERIN”COLOUR INDICATORS IRRADIATED IN FIXED POSITION

Dose (Gy)	STERIN-125	STERIN-300
0	n.a.	n.a.
50	+	n.a.
100	++	n.a.
150	++	+
200	++	+
300	n.d.	++
500	n.d.	++

() “NOT” could be seen clearly; (+) “NOT” could hardly be seen; (++) “NOT” is completely covered; (n.a.) not applicable (n.d.) not determined.

3.2.3. Irradiation in commercial practice

Figs and raisins were irradiated during normal processing at the UTR facility to simulate/validate a commercial practice.

Table 5 shows the dose uniformity data by using the Harwell Amber Perspex dosimeter and Clear Perspex dosimeter prepared at the irradiation facility. The dosimeters were placed as indicated in Figure 1. The average dose uniformity was 1.38 for the Harwell Amber Perspex dosimeter and 1.35 Clear Perspex dosimeter.

TABLE 5. DOSE DISTRIBUTION STUDIES BY USING HARWELL AMBER PERSPEX AND CLEAR PERSPEX DOSIMETERS

Dosimeters	Dried Fruits: figs, raisins	Absorbed Dose (kGy) $Dose_{min}$	Absorbed Dose (kGy) $Dose_{max}$	Average Dose Uniformity
Harwell Amber Perspex (651 nm)	Box 1	14.0	19 2	1.38
	Box 2	14.3	19.9	
Clear Perspex (390 nm)	Box 1	16.5	22.0	1.35
	Box 2	17.3	23.6	

4. CONCLUSIONS

The parameters to irradiate the dried fruits selected for this study were established. The location of minimum absorbed dose and maximum absorbed dose were determined. To a Dmin defined in the central plan of each UTR box corresponds a Dmax at its external faces, which is not higher enough to damage the fruits. The dose uniformity was acceptable for an industrial gamma irradiation facility.

The label indicators utilised in this study needs to be more explored but our results indicate that it works well on the interval of doses used for disinfestation.

Further work shall be done with the Clear Perspex. Other thickness 5 mm and 8 mm adequate to lower absorbed dose shall also be tested.

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REFERENCES

[1] FARKAS, J., Reducing microbial populations in dry ingredients by ionising radiations, in Iradiation of dried food ingredients (1988), CRC Press, Inc., Boca Raton, Florida.

[2] ICGFI DOCUMENT No. 20, Code of Good Irradiation Practice for Insect Disinfestation of Dried Fruits and Tree Nuts, ICGFI, Vienna (1994).

[3] ANDRADE, M.E., POLÓNIA, I., Establishment of Technological Parameters to Irradiate Dried Figs (1996), Second Research Coordination Meeting on "Standardized Methods to Verify Absorbed Dose in Fresh and Dried Fruits, Tree Nuts in Trade", Federal Research Centre for Nutrition, Karlsruhe, 59 August.

[4] MENDES, C.M., ALMEIDA, J.C., BOTELHO, M.L., CAVACO, M.C., ALMEIDAVARA, E., ANDRADE, M.E., The portuguese Irradiation Facility, Phys. Chem. **35** (46) (1990) 576–579.

[5] CAVACO, M.C., ALMEIDA, J.C., ANDRADE, M.E., KOVÀCS, A., Appl. Radiat. Isot. **42** (12) (1991) 1185–1188.

[6] ANDRADE, M.E., COELHO, N., OLIVEIRA, J.E., Upgrading of a Gamma Radiation Facility, Radiat. Phys. Chem. **46** (1990) 503–506.

[7] E102692. Practice for Using the Fricke Reference Standard Dosimetry System. In: Annual Book of ASTM Standards, 12.02 (1992) 620–626, ASTM, Philadelphia.

[8] CHADWICK, K.H., The Effect of Light Exposure on the Optical Density of Irradiated Clear Polymethylmethacrylate, Phys.Med.Biol. **17** (1) (1972) 88–93.

[9] MCLAUGHLIN, W.L., BOYD, A.W., CHADWICK; K.H., MCDONALD, J.C., MILLER, A., Dosimetry for Radiation Processing (1989), Taylor and Francis, London.

[10] EHLERMANN, D.A.E., Validation of a Label Dosimeter for Food Irradiation Applications by Subjective and Objective Means, Radiat. Phys. Chem. **48** (1997) 1197–1201.

[11] RAZ, M.D., Dosimetric Performance of and Environmental Effects on Sterin Irradiation Indicators Labels, Radiat. Phys. Chem. **49** (1997) 491–495.

DEVELOPMENT OF PROCESS CONTROL FOR THE IRRADIATION OF FRESH MANGOES

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Abstract. Dose distribution studies in mangoes contained in boxes used in commercial trade for export, were done using the multipurpose irradiation facility at the Philippine Nuclear Research Institute. The mangoes were irradiated at a target dose of 100 Gy, the dose required for quarantine treatment of fresh mangoes against fruitflies. Positions of minimum dose and maximum dose were identified and dose uniformity ratio was determined. Fricke and Gammachrome YR dosimeters were used for the dose distribution studies. The performance of STERIN threshold indicators was evaluated by irradiating them at different doses. STERIN 125 indicators were also attached to the surface of the mango boxes during the dose distribution studies. STERIN indicators can be useful to differentiate between irradiated and unirradiated products.

1. INTRODUCTION

The "Carabao" (Manila Super) mango has a good export potential and is an important dollar earner for the Philippines. However, because of the presence of fruitflies in the Philippines, most importing countries would require quarantine treatment for this commodity. Japan for example, requires vapor heat treatment (VHT) as the method of disinfestation for mangoes imported from the Philippines.

A comparative study [1] between radiation and vapor heat treatment showed that irradiation at a dose of 100 or 250 Gy has no adverse effect on fruit quality. Irradiation offers a distinct advantage over VHT because it does not induce internal breakdown, while VHT can induce a physiological disorder in mangoes, characterized by the presence of unsightly and tough spongy tissues.

Using emergence of adult fruitflies as the standard of survival, it has been established [2, 3] that a dose of 100 Gy meets the Probit 9 (99.9968%) level of mortality as a basis of quarantine security.

Since effectiveness of the radiation treatment is very much dependent on the dose received by the product, the present study therefore aims to develop process control for the irradiation of fresh mangoes as quarantine treatment against fruitflies.

2. METHODS

2.1. Variation of bulk density of mangoes

A major exporter of mangoes was visited. Operations in their plant were observed. Special attention was given to sorting, sizing and packaging. Mangoes used in this study were obtained from this exporter. Variation of bulk density of mangoes contained in boxes used for export was determined.

2.2. Dosimeter

2.2.1. Fricke dosimeter

Fricke dosimeter was prepared according to standard procedure [4]. A Jasco Model 7800 spectrophotometer was used to determine the absorbance of the Fricke dosimeters. The wavelength of this spectrophotometer was checked using holmium and didymium glass filters, while the absorbance was checked using an absorbance standard set, both from Pye Unicam Ltd.

2.2.2. Gammachrome YR dosimeter

Gammachrome YR (polymethylmethacrylate) dosimeters were used as obtained from Harwell. They were calibrated [5] using Fricke dosimeter as the reference. Three (3) dosimeters each at eight (8) dose levels were irradiated at a specific position in a Gammacell 220, whose dose rate has been previously determined by Fricke dosimeter.

The absorbance of the irradiated Gammachrome dosimeters was measured using a Jasco Model 7800 spectrophotometer at 530 nm, two hours after irradiation. The thickness of the dosimeters was measured by a Mitutoyo Digimatic caliper, whose response was checked against a Mitutoyo standard gauge block. For the calibration curve, a third order polynomial was fitted to the data to get the relationship between specific absorbance and dose.

2.3. Dose distribution in product boxes

Irradiation of mangoes was done at the multipurpose irradiation facility of the Philippine Nuclear Research Institute (PNRI). This irradiation facility is a batch type Gammabeam 651PT from Nordion International with a total loading about 5.6 PBq ^{60}Co.

Green mangoes, contained in boxes, were loaded on turntables, which were located 56 cm from the source plaque and 48 cm from the center line of the source (Fig. 1). Loading on one turntable consisted of 2 stacks of 6 boxes each (a total of 12 boxes). Product boxes were placed on the turntables such that the midplane and midheight of the product loading coincided with the center of the turntable and the symmetry level of the source plaque respectively. The turntables were made to rotate (4 quarter turns) during the irradiation period. The mangoes were irradiated to a target dose of 100 Gy, the dose required for quarantine treatment against fruitflies.

All trials were carried out with mangoes of medium size. For each trial, only turntables 1 and 4, which are located on one side of the source plaque, were loaded with mangoes, since results of previous studies [6] showed that due to symmetry, dose distribution on the other side of the source plaque would be similar. The mangoes were contained in boxes, 46 × 32 ×14 cm. Inside these boxes are partitions, which separate individual mangoes from each other. Each box contained about 5 kg of mangoes. For medium size mangoes, there were 20 mangoes per box.

To determine the dose distribution in the product boxes, Fricke dosimeters were placed inside the boxes, one in each partition as shown in Fig. 2. The positions of minimum dose and maximum dose, as well as dose uniformity ratio were determined. Dosimeters were also placed at reference positions, located outside the box at the center of plane I.

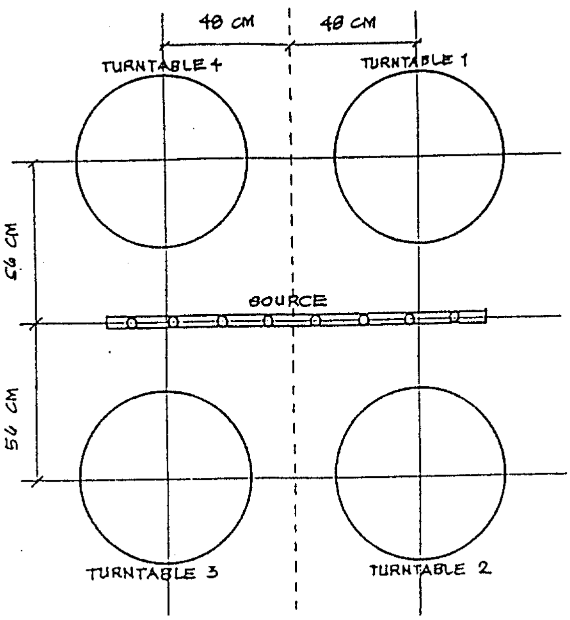

FIG. 1. Position of turntables relative to source.

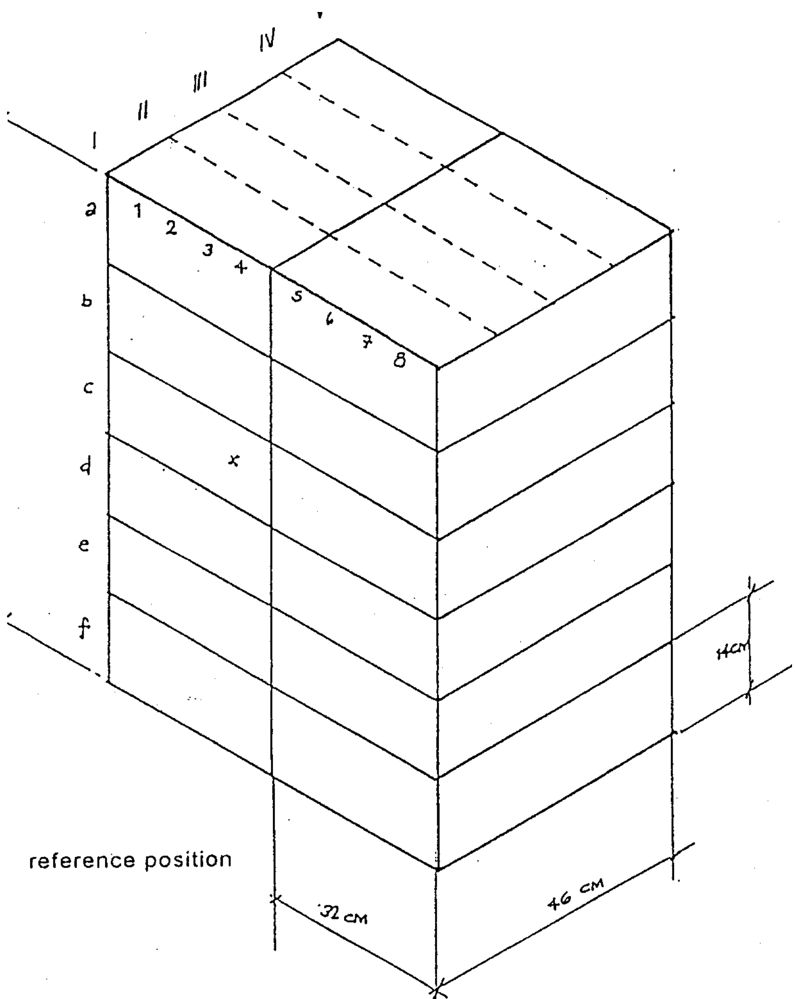

FIG. 2. Positions of dosimeters in mango boxes.

Subsequently, in the other trials, dosimeters were placed only at strategic positions to monitor the consistency of the minimum and maximum dose and their positions.

To compare the response of the Fricke and Gammachrome dosimeters, they were placed together in some critical positions in the product boxes.

2.4. Threshold dose indicators

Threshold indicators, STERIN 125 and STERIN 300 were received from International Specialty Products [7]. STERIN 125 is a threshold indicator for 125 Gy while STERIN 300 is for 300 Gy. The threshold indicators were irradiated at different doses, in a Gammacell 220, whose dose rate has been determined by Fricke dosimeter.

Color measurements on the irradiated indicators were done using a spectrocolorimeter (SZ80 II Color Measuring System, Nippon Denshoku, Kogyo Co. Ltd, Japan). Color of the samples were expressed in terms of the following systems of color specification:

(a) CIE (Commission Internationale d'Eclairage) Color System using:

– Tristimulus values: XYZ
– Lightness value and chromaticity coordinates: Yxy

(b) Hunter Color System using Hunter Lab Values and measurement of ΔE (total color difference) using unirradiated sample as reference.

Visual examinations to determine presence/absence of the word "NOT" in the irradiated indicators were conducted by trained/experienced laboratory panelists.

STERIN 125 indicators were also attached to the outside surface of the mango boxes, near the reference positions. Visual changes in the indicators were evaluated.

2.5. Infestation of mangoes

Mangoes were infested with Oriental fruit fly eggs, by inoculating twentyfour to twentyfive hour old eggs into tiny holes made by puncturing the mango fruits with a "frog". About 150 eggs were inoculated into each fruit. For each trial, four infested mangoes were placed at the expected minimum dose position and irradiated together with the uninfested mangoes. Four control mangoes were also infested with the same number of eggs.

Both irradiated and unirradiated infested fruits were held in plastic jars containing coir dust for about two weeks, after which the fruits were observed for pupal formation. The coir dust was sieved and the fruits dissected to gather pupae. The pupae were held in plastic cups until emergence. Normal flies were counted and recorded as survivors.

3. RESULTS AND DISCUSSION

3.1. Variation of bulk density of mangoes

Mangoes are sorted according to weight per fruit and packed according to size as shown in Table I. Mangoes for export are packed in corrugated carton boxes, 46 × 32 × 14 cm. Each box contains 5 kg of mangoes.

TABLE I. SIZES OF MANGOES

Size	Weight per piece, (g)	Number of pieces per box
Extra small, SS	150 to 199	30
Small, S	200 to 229	24
Medium, M	230 to 269	20
Large, L	270 to 349	16
Extra large, LL	350 up	12

TABLE II. WEIGHT OF MANGO BOXES

	Trial 1	Trial 2	Trial 3	Trial 4	Trial 5
Ave. weight, kg	5.85	5.76	6.02	5.82	5.80
SD	0.22	0.09	0.24	0.11	0.13
CV (%)	3.8	1.6	4.0	1.9	2.2
Min. weight, kg	5.5	5.6	5.6	5.6	5.6
Max. weight, kg	6.2	5.9	6.4	6.0	6.1
Min. density, kg/m^3	267	272	272	272	272
Max. density, kg/m^3	301	286	311	291	296

Size of mangoes: medium
n = 24 boxes per trial

Overall ave. weight: 5.85 kg
Overall ave. density: 284 kg/m^3

Table II shows the variation in gross weights of packed mangoes. Balance used for weighing has a readability of 0.1 kg. Overall average weight per box was 5.85 kg, while individual box weights ranged from 5.5 to 6.4 kg. Thus bulk density varied from 267 kg/m^3 to 311 kg/m^3.

3.2. Dose distribution in product boxes

Since results of dose distribution showed that the overall minimum absorbed dose was at the central plane on turntable 1, while overall maximum dose was at plane I on turntable 4, the loading on both turntables 1 and 4 can be considered as a “process unit”. The frequency distribution and the normal probability plot of dose of the process unit are shown in Fig. 3 and 4 respectively. It can be observed that the frequency distribution of dose of the process unit approaches a normal distribution, with average dose equal to 145 Gy and standard deviation equal to 18 Gy. Minimum dose was 100 Gy while maximum dose was 197 Gy. It can also be shown that a rough estimate of the average dose can be obtained by averaging the minimum and maximum doses.

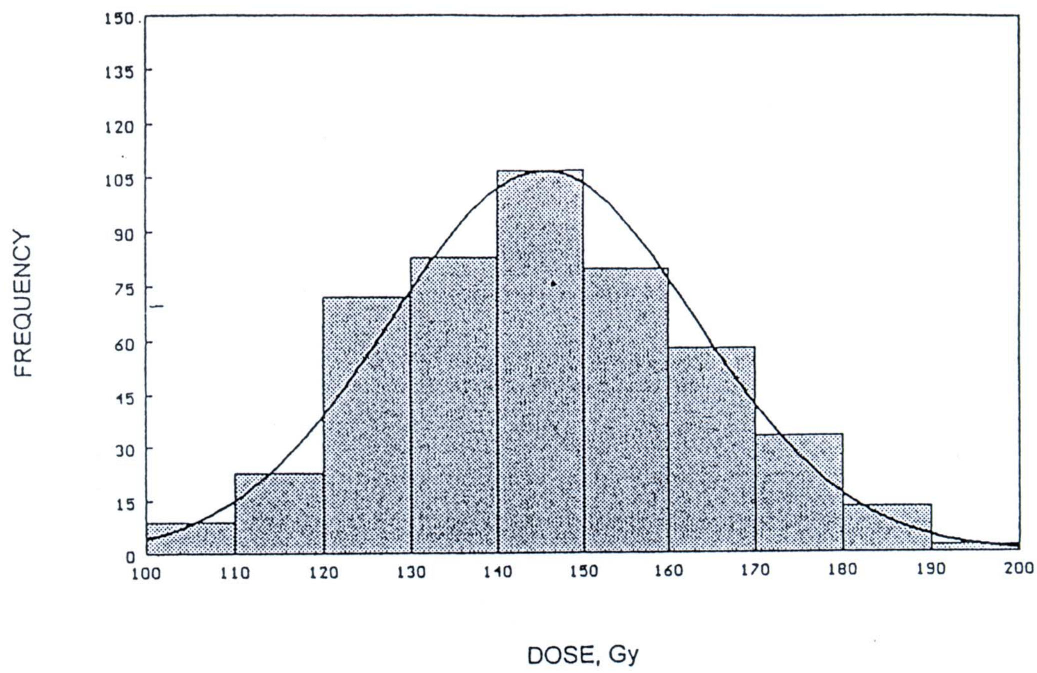

FIG. 3. Frequency distribution of dose on a process unit (turntables 1 and 4).

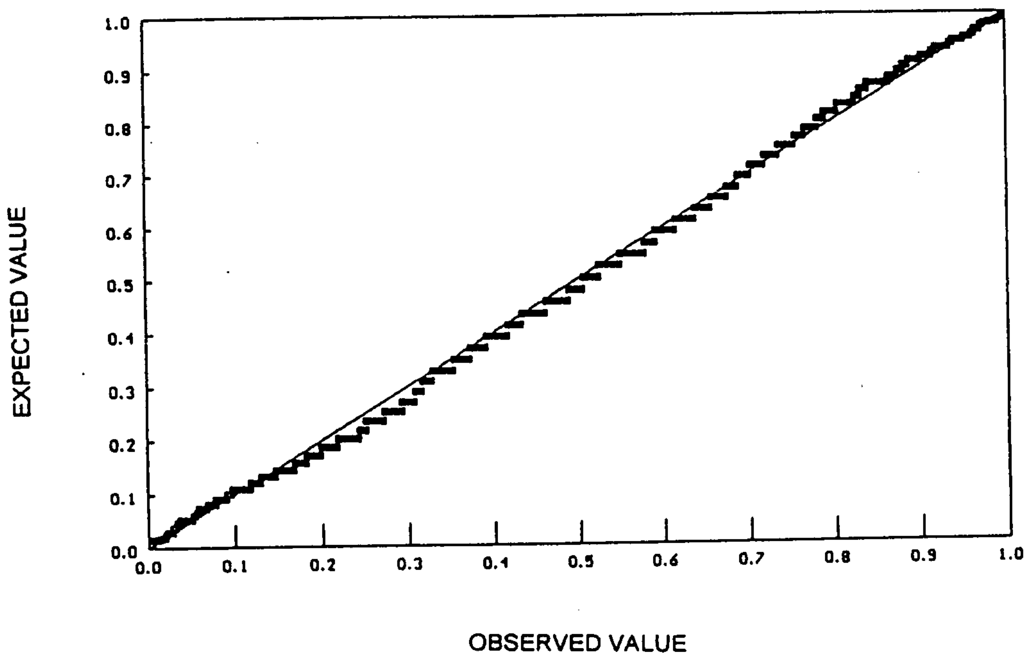

FIG. 4. Normal probability plot of dose on a process unit (turntables 1 and 4).

Results of several trials showed that the minimum absorbed dose was at the central plane at position III5f or III4f on turntable 1 while maximum dose was on plane I at position I1d on turntable 4. Table III shows the minimum and maximum dose as well as dose uniformity ratio obtained from several trials. Similar results for minimum dose, maximum dose and dose uniformity ratio were obtained in all trials. It is worth noting that the different trials were made over a span of about two years. Conditions for irradiation were similar except for time of irradiation, which was corrected to account for decay of the source.

The relationship between the dose at two reference positions (position A on turntable 1 and position B on turntable 4) and the minimum and maximum doses are also shown in Table III. Again similar results were obtained from all trials.

TABLE III. MINIMUM DOSE, MAXIMUM DOSE AND DOSE UNIFORMITY RATIO

	Trial 1	Trial 2	Trial 3	Trial 4	Trial 5
Minimum Dose	103	102	100	106	111
Maximum Dose	185	187	197	189	195
Dose uniformity ratio	1.81	1.83	1.97	1.78	1.76
Dose at ref A	150	149	149	157	155
Dose at ref B	177	170	176	174	176
Min dose/ref A	0.69	0.68	0.67	0.68	0.72
Min dose/ref B	0.58	0.60	0.57	0.61	0.63
Max dose/ref A	1.23	1.26	1.32	1.20	1.26
Max dose/ref B	1.05	1.1	1.12	1.09	1.11

same source geometry
time of irradiation corrected for decay
minimum dose position: bottom of central plane (III4f or III5f) on turntable 1
maximum dose position: edge of plane I (I1d) on turntable 4
reference position A: outside on center of plane I on turntable 1
reference position B: outside on center of plane I on turntable 4

3.3. Gammachrome YR dosimeter

In 2 trials, Gammachrome dosimeters were placed together with the Fricke dosimeters in some critical positions of the mango boxes. Table IV shows results of this comparison. It is observed that on the average Gammachrome gave dose readings slightly higher than Fricke dosimeters. It should be noted that the dose range for quarantine treatment of mangoes is at the lower end of the recommended dose range for Gammachrome (100 Gy to 3 kGy).

TABLE IV. COMPARISON BETWEEN FRICKE AND GAMMACHROME

	Trial 1			Trial 2	
Fricke (1)	Gammachrome (2)	Ratio (2):(1)	Fricke (3)	Gammachrome (4)	Ratio (4):(3)
195	188	0.96	174	198	1.14
176	194	1.10	188	207	1.10
186	187	1.01	184	201	1.09
129	117	0.91	106	102	0.96
128	120	0.94	179	213	1.19
186	210	1.13	189	227	1.20
186	199	1.07	157	147	0.94
177	191	1.08	161	178	1.11
155	162	1.05	108	108	1.00
163	174	1.07	167	198	1.19
111	86	0.78	161	179	1.11
168	176	1.05	123	129	1.05
172	193	1.12	171	205	1.20
			122	121	0.99
	Ave.	1.02		Ave.	1.09
	CV(%)	9.7		CV(%)	8.3

TABLE V. COLOR MEASUREMENT (USING CIE COLOR SYSTEM) OF IRRADIATED STERIN 125

Dose Gy	Sample Code	Spectrocolorimeter Reading [a] Color Specification					
		Y	X	Z	Y	x	y
0	O1	13.93	17.19	12.39	13.93	0.3950	0.3201
	O2	13.92	17.24	12.38	13.92	0.3959	0.3197
20	17	11.11	11.09	10.21	11.11	0.3421	0.3427
	18	11.02	10.91	10.03	11.02	0.3413	0.3448
40	5	8.76	6.93	8.68	8.76	0.2843	0.3594
	6	8.92	7.03	8.89	8.92	0.2830	0.3590
60	15	8.49	5.83	8.58	8.49	0.2545	0.3707
	16	8.49	5.88	8.57	8.49	0.2563	0.3700
80	3	7.77	5.01	8.22	7.77	0.2385	0.3699
	4	7.81	4.98	8.23	7.81	0.2369	0.3715
90	13	7.92	5.04	8.34	7.92	0.2366	0.3718
	14	7.77	4.89	8.26	7.77	0.2337	0.3714
100	7	7.80	4.85	8.21	7.80	0.2324	0.3739
	8	7.82	4.87	8.21	7.82	0.2330	0.3741
105	23	7.81	4.88	8.33	7.81	0.2321	0.3715
	24	7.83	4.89	8.33	7.83	0.2322	0.3719
110	19	7.70	4.75	8.12	7.70	0.2309	0.3743
	20	7.74	4.76	8.16	7.74	0.2303	0.3746
115	9	7.74	4.75	8.22	7.74	0.2293	0.3737
	10	7.81	4.84	8.30	7.81	0.2310	0.3727
120	1	7.49	4.56	7.91	7.49	0.2284	0.3752
	2	7.49	4.57	7.95	7.49	0.2283	0.3742
125	11	7.41	4.50	7.91	7.41	0.2270	0.3738
	12	7.46	4.58	8.10	7.46	0.2274	0.3704
130	21.	7.34	4.50	8.02	7.34	0.2265	0.3695
	22	7.38	4.49	7.97	7.38	0.2263	0.3719

[a]Values average of 3 measurements.

TABLE VI. TOTAL COLOR DIFFERENCE (ΔE, HUNTER COLOR SYSTEM) OF IRRADIATED STERIN 125

Dose Gy	Sample Code	Spectrocolorimeter Reading [a] Color Specification L	a	b	Total Color Difference (ΔE)[b]
80	3	27.87	16.70	2.05	35.16
	4	27.94	17.10	2.12	35.51
90	13	28.14	17.28	2.15	35.64
	14	27.87	17.47	1.96	35.91
100	7	27.92	17.88	2.14	36.26
	8	27.96	17.85	2.19	36.22
105	23	27.94	17.74	1.91	36.15
	24	27.98	17.78	1.96	36.17
110	19	27.74	18.01	2.09	36.44
	20	27.82	18.15	2.10	36.55
115	9	27.82	18.21	1.98	36.63
	10	27.94	17.99	1.97	36.39
120	1	27.36	18.15	2.04	36.69
	2	27.36	18.09	1.95	36.64
125	11	27.22	18.13	1.85	36.73
	12	27.31	17.87	1.56	36.49
130	21	27.09	17.76	1.43	36.47
	22	27.16	18.04	1.64	36.68

[a] values are average of 3 measurements.
[b] lab values of reference sample (0 Gy).

L = 37.32 a = 16.88 b = 6.47

3.4. Threshold dose indicator

Results of color measurements (CIE color system) of STERIN 125 irradiated at different doses are shown in Table V while total color difference (ΔE) under the Hunter Color System, using the unirradiated sample as reference are shown in Table VI.

Results in Table V and VI seem to indicate that quantifying the response of the STERIN125 by spectrocolorimeter may not be practical since there is no obvious relationship between dose and color specification.

Results of visual examination by trained panelists (Table VII.A) indicate that STERIN 125 starts to show visual changes, i.e. absence of the word “NOT” at 125 Gy (6 out of 12 judgements). This observation agrees with the claim of the supplier that the word “NOT” becomes obscure at a dose of 125 Gy and above.

A paired comparison test (Table VII.B) shows that there is a significant difference in appearance (color and intensity of the word “NOT”) between samples irradiated at 120 and 125 Gy. Again this agrees with the supplier’s claim that STERIN 125 is a threshold indicator for 125 Gy.

All STERIN 125 indicators attached to the surface of mango boxes, which were irradiated at a target dose of 100 Gy showed the required response, i.e. the word “NOT” became obscured, indicating that the product was “IRRADIATED”.

TABLE VII. VISUAL EXAMINATION OF IRRADIATED STERIN 125

A. Visual examination to determine absence of the word “NOT”

Dose Gy	Code	No. of judgments indicating absence of word “NOT”
80	3	0 out of 12 judgments
90	14	0 out of 12 judgments
100	7	0 out of 12 judgments
105	23	0 out of 12 judgments
110	19	0 out of 12 judgments
115	9	0 out of 12 judgments
120	1	2 out of 12 judgments
125	11	6 out of 12 judgments
130	21	6 out of 12 judgments

B. Paired comparison test for difference

Samples: 120 Gy (Code 2) and 125 Gy (Code 11)
Type of analysis: binomial test
Level of significance: 5%

Sensory Attribute	Number of Correct Judgment	Significance (based on number of correct judgment)
Color	10	with significant difference
Intensity of word “NOT”	10	with significant difference

Conditions for both evaluations:
Conducted by 6 trained/experienced laboratory panelists (2 replications/panelist or total of 12 judgments)
Order of Presentation: Randomized
Light Source: Four pieces of fluorescent bulbs (Philips SL, 18 watts), covered with diffuser, located 3.7 feet above the sample
Viewing Condition: Sample laid horizontally using black illustration board as background and viewed at an angle of approximately 45°

On the other hand, results of color measurements (CIE color system) of STERIN 300 irradiated at different doses are shown in Table VIII and total color difference (Hunter Color System) are shown in Table IX.

Similar to STERIN 125, results in Table VIII and IX seem to indicate that quantifying the response of the STERIN 300 by spectrocolorimeter may not be practical since there is no obvious relationship between dose and color specification.

TABLE VIII. COLOR MEASUREMENT (USING CIE COLOR SYSTEM) OF IRRADIATED STERIN 300

Dose Gy	Sample Code	Spectrocolorimeter Reading [a]					
		Color Specification					
		Y	X	Z	Y	x	y
0		19.59	25.21	16.26	19.59	0.4128	0.3208
250	a	9.14	5.87	9.92	9.14	0.2354	0.3666
	b	9.10	5.80	9.89	9.10	0.2339	0.3670
270	c	8.99	5.80	10.01	8.99	0.2338	0.3624
	d	9.02	5.75	10.03	9.02	0.2318	0.3637
290	g	9.05	5.78	10.04	9.05	0.2324	0.3638
	h	9.05	5.77	10.04	9.05	0.2320	0.3640
300	k	9.01	5.75	10.03	9.01	0.2319	0.3634
	l	9.07	5.79	10.10	9.07	0.2319	0.3633
310	o	8.97	5.69	9.92	8.97	0.2314	0.3649
	p	8.99	5.75	10.14	8.99	0.2310	0.3613
320	s	8.90	5.55	9.87	8.90	0.2281	0.3659
	t	9.06	5.65	9.94	9.06	0.2292	0.3675
330	w	8.84	5.50	9.78	8.84	0.2280	0.3664
	x	8.82	5.47	9.87	8.82	0.2263	0.3650
340	aa	9.01	5.63	10.10	9.01	0.2275	0.3641
	bb	8.69	5.42	9.69	8.69	0.2272	0.3643
360	ee	8.99	5.62	10.13	8.99	0.2271	0.3633
	ff	8.93	5.60	10.20	8.93	0.2264	0.3610
365	pp	9.11	5.70	10.26	9.11	0.2273	0.3633
	qq	9.10	5.69	10.26	9.10	0.2271	0.3632
370	rr	9.13	5.69	10.05	9.13	0.2287	0.3670
	ss	8.99	5.57	9.90	8.99	0.2277	0.3675
380	nn	9.07	5.62	9.96	9.07	0.2279	0.3679
	oo	9.06	5.63	10.00	9.06	0.2280	0.3669
390	mm	9.04	5.63	9.94	9.04	0.2287	0.3673
	tt	9.06	5.63	10.09	9.06	0.2271	0.3656
400	jj	8.85	5.43	10.00	8.85	0.2236	0.3644
	uu	9.09	5.55	10.13	9.09	0.2240	0.3669

[a]Values average of 3 measurements.

TABLE IX. TOTAL COLOR DIFFERENCE (ΔE, HUNTER COLOR SYSTEM) OF IRRADIATED STERIN 300

Dose Gy	Sample Code	Spectrocolorimeter Reading [a] Color Specification L	a	b	Total Color Difference (ΔE)[b]
250	a	30.23	18.25	1.73	45.32
	b	30.16	18.47	1.70	45.56
270	c	29.98	17.94	1.22	45.20
	d	30.03	18.39	1.25	45.59
290	g	30.08	18.35	1.29	45.54
	h	30.08	18.41	1.29	45.59
300	k	30.01	18.34	1.22	45.56
	l	30.11	18.39	1.22	45.57
310	o	29.94	18.50	1.35	45.71
	p	29.98	18.24	0.96	45.52
320	s	29.83	19.00	1.29	46.22
	t	30.09	19.17	1.51	46.26
330	w	29.73	19.01	1.33	46.26
	x	29.69	19.10	1.11	46.39
340	aa	30.01	19.05	1.09	46.25
	bb	29.47	18.77	1.07	46.16
360	ee	29.98	19.01	0.98	46.24
	ff	29.88	18.85	0.70	46.17
365	pp	30.18	19.11	1.00	46.27
	qq	30.16	19.12	0.97	46.29
370	rr	30.21	19.27	1.45	46.33
	ss	29.98	19.31	1.43	46.44
380	nn	30.11	19.40	1.50	46.47
	oo	30.09	19.29	1.40	46.39
390	mm	30.06	19.19	1.47	46.30
	tt	30.09	19.29	1.22	46.42
400	jj	29.74	19.48	0.92	46.76
	uu	30.14	19.90	1.21	46.99

[a] values are average of 3 measurements.
[b] lab values of reference sample (0 Gy).
L = 44.26 a = 24.19 b = 9.23

Results of visual examination by trained panelists (Table X) indicate that STERIN 300 starts to show visual changes, i.e. absence of the word "NOT" at 380 Gy (6 out of 12 judgements). However the supplier claims that for STERIN 300, the word "NOT" becomes obscure at a dose of 300 Gy and above. This discrepancy may due to the fact that the STERIN 300 were irradiated one month before its expiration date.

TABLE X. VISUAL EXAMINATION OF IRRADIATED STERIN 300

Dose Gy	Code	No. of judgments indicating absence of word “NOT”
250	a	0 out of 12 judgments
270	c	0 out of 12 judgments
290	g	0 out of 12 judgments
300	k	0 out of 12 judgments
310	p	0 out of 12 judgments
320	t	0 out of 12 judgments
330	w	0 out of 12 judgments
340	aa	0 out of 12 judgments
360	ee	0 out of 12 judgments
365	qq	4 out of 12 judgments
370	l	4 out of 12 judgments
380	nn	6 out of 12 judgments
390	tt	6 out of 12 judgments
400	uu	12 out of 12 judgments

Conditions for both evaluations:
Conducted by 6 trained/experienced laboratory panelists
(2 replications/panelist or total of 12 judgments)
Order of Presentation: Randomized
Light Source: Four pieces of fluorescent bulbs (Philips SL, 18 watts), covered with diffuser, located 3.7 feet above the sample
Viewing Condition: Sample laid horizontally using black illustration board as background and viewed at an angle of approximately 45°

3.5. Infestation of mangoes

As shown in Table XI, most of the irradiated eggs of Oriental fruit flies could not transform to the pupal stage. None of the recovered pupae from irradiated eggs emerged into adult. This confirmed results of previous studies, which showed that a dose of 100 Gy is sufficient as quarantine treatment for Oriental fruit flies.

TABLE XI. FRUIT FLY EMERGENCE FROM INFESTED MANGOES

Treatment/ Replications	Trial 1 Pupae Recovery	Emerged Adult	% Adult Emergence	Trial 2 Pupae Recovery	Emerged Adult	% Adult Emergence
Control						
1	69	32	46.38	28	18	64.28
2	67	43	64.18	45	41	91.11
3	55	38	69.09	18	12	66.67
4	78	23	29.49	65	45	69.23
Irradiated						
1	0			0		
2	0			0		
3	2	0	0	0		
4	2	0	0	1	0	0

150 eggs infested per fruit.

4. CONCLUSION

These studies confirmed that irradiation of fruit fly eggs at a dose of 100 Gy results in no adult fruit fly emergence.

The frequency distribution of dose is almost normal. A rough estimate of average dose can be obtained by averaging the minimum and maximum dose.

The dose distribution studies showed where the positions of minimum dose and maximum dose are located. These positions are to be monitored during routine irradiation, as a process control, to confirm that indeed the products have received at least the minimum dose required and not more than the maximum dose, which in some instances may have deleterious effects on the product. In cases when the minimum and maximum dose positions are not readily accessible, the dose at a reference position can be used instead to monitor the dose received by the product. However the relationship between dose at the reference position and minimum dose and the relationship between dose at the reference position and maximum dose should first be established.

STERIN 125 is a convenient indicator, which provides a visual verification that the product has been irradiated at or above 125 Gy. Though not a dosimeter, STERIN 125 can be used to differentiate between irradiated and unirradiated products.

ACKNOWLEDGEMENTS

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REFERENCES

[1] LIZADA, M.C.C., et al., Comparative Effects of Gamma Radiation and the Vapor Heat Treatment on Fruit Quality of "Carabao" (Manila Super) Mangoes, Phil. Nuclear J. **7** (1990) 919.

[2] MANOTO, E.C., et al., Effects of Gamma Radiation on Mortality of the Oriental Fruit Fly, *Dacus Dorsalis Hendel*, when Inoculated in Manila Super Mango, Phil. Nuclear J., **6** (1989) 453–458.

[3] MANOTO, E.C., et al., Irradiation as a Quarantine Treatment of "Carabao" (Manila Super) Mangoes, Phil. Nuclear J. **7** (1990) 17.

[4] Standard Practice for Using the Fricke Reference Standard Dosimetry System, E102692, ASTM Vol. 12.02 (1994).

[5] Standard Practice for Use of a Polymethylmethacrylate Dosimetry System, E127693, ASTM, Vol. 12.02 (1994).

[6] CABALFIN, E.G., et al., Dose Mapping of the Multipurpose Gamma Irradiation Facility, Phil. Nuclear J. **6** (1989) 416–129.

[7] Data Sheet, Sterin Irradiation Indicators, International Specialty Products.

EVALUATION OF A LABEL DOSIMETER TO BE USED FOR BRAZILIAN IRRADIATED FRESH FRUITS

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Abstract. The main difficulties for Brazilian fruit exports are phitosanitary barriers. Irradiation can be used as a single treatment, part of a multiple treatment or combined with other mitigation measures as a component of a systems approach which would be a treatment for plant pests of quarantine significance. For any kind of industrial irradiation, determining the absorbed doses involves a dosimetry system that covers the absorbed dose range of interest and shall be calibrated before use. Frequently, however, it is useful to also have a radiation sensitive indicator to visually determine whether or not a product has been irradiated. STERIN labels were designed as threshold indicators, where a visual message changes after exposure at or above the threshold indication dose (e.g. 125 Gy, 300 Gy). The aim of this work was to evaluate STERIN label indicators to be used for Brazilian irradiated fresh fruits.

1. INTRODUCTION

1.1. Irradiation as a quarantine treatment

Radiation processing is a rapidly developing technology with several applications in the field of food preservation. Irradiation can be used as a single treatment, part of a multiple treatment or combined with other mitigation measures as a component of a systems approach for quarantine purposes. Despite the fact that irradiation cannot be considered the only replacement for methyl bromide, it is recognized that there is adequate scientific evidence to prove that irradiation provides an alternative treatment to be explored and developed, with potentially broad applications in the treatment of quarantine plant pests.

Measures aimed at reducing pest presence prior to treatment must be encouraged but are not required for quarantine security treatments like irradiation. However, a very low initial infestation rate is important for enhancing the acceptance and use of irradiation as a treatment and for alleviating regulatory concerns deriving from the detection of living pests in the irradiated product. In those instances where pest organisms survive treatment, it is essential, for quarantine purposes, that the organism be unable to reproduce, and it is desirable for the organism to be unable to emerge from the commodity unless it can be easily distinguished from a nonirradiated pest of the same species.

The irradiation can be applied to bulk or continuous unpacked commodities, as an integral part of packing operations. It may be done at a central location such as the port of embarkation after packing or packaging. It may also be performed at the port of arrival or a designated location in the destination country when safeguards are deemed to be adequate and operationally feasible.

Irradiation treatment must be carried out to ensure that the minimum absorbed dose required to assure quarantine security is fully attained throughout the commodity. The schedule process for the minimum absorbed dose must account for uncertainty associated with the dosimetry system employed. The maximum dose may also be required in order to comply with national requirements for some commodities. Also, a dose mapping of the product in every geometric packing configuration, arrangement and product density that will be used during routine treatments will be required prior to the approval of the facilities. Dose and dose distribution are determined by product parameters and by source parameters. Products parameters are primarily the density of the food itself and the density of packing the individual food containers within the tote box or carrier in which irradiation takes place.

Source parameters are different for the different types of irradiators. In the case of gamma irradiators, the relevant factors are the isotope, source strength and geometry, source pass configuration and mechanism, conveyor speed and dwell time. In the case of machine sources, the relevant factors are type of radiation (electron beam or X rays), beam energy and beam power (MeV and kW), scan width and scan frequency, pulse repetition rate in the case of pulsed electron beams, beam pass configuration and mechanisms and conveyor speed. Some of these factors are constant for a given irradiator (e.g. type of isotope or radiation, design geometry), others change systematically (source strength), and still others can be set by an operator according to the requirements of the process (e.g., dwell time).

An International Task Force on Irradiation as a Quarantine Treatment of Food and Agricultural products convened by the ICGFI in 1991, prepared a Code of Good Irradiation Practice for Insect Disinfestation of Fresh Fruits [1]. The document established the absorbed radiation doses required to provide treatment of fruits to meet quarantine criteria for fruit fly and other pests of major international economic and quarantine importance.

Based on the thorough work performed worldwide to show the efficiency of irradiation for disinfestation and the capability of the technique to be used as a quarantine treatment, the US Federal Register published the policy statement concerning the use of irradiation as a treatment for plants pests of quarantine significance on May 15, 1996. As the USA is an important importer of fruits, the launching of this regulatory document is an important landmark in the applicability of the technique.

1.2. Considerations about Brazilian fruits

Brought by the Portuguese at the beginning of the XVIth century, citric fruits (*Citrus spp*) coming from the East adjusted well to Brazilian soils. Today, the country is the world's largest producer of fruits (bananas, mangoes, melons, papayas, grapes, apples, guavas, pineapples, figs) and especially citrus output at 19.7 million tons (mt) in 1997, an increase of 12 per cent over the previous year. For the present harvest, orange production is forecasted to reach a record 18.3mt (source USDA). More than half of the total citrus output is destined for processing, particularly for frozen juice concentrate; 25 per cent is destined to the domestic market. Only a small percentage is earmarked for export: from a production of 31mt, 725,000 tons are exported. In the case of oranges, for example, this is a little more than five per cent of the total fresh output. Brazilian mango exports increased 360% from 1989 to 1996, from 5,400t to 24,200t. Even considering these figures, Brazilian exports of fresh fruits in 1996 were no more than US$ 104 m. The majority of fruits destined to Europe is exported to the Netherlands, though gains are being made in the UK and France. However, shipping to the United States has declined, largely because Brazil has strengthened its position on other markets, notably Canada, Japan and Korea [2].

The citrus production is concentrated in São Paulo where output is increasing as new trees come on stream. Recently, however, the NorthEast region of the country (mainly the States of Bahia and Sergipe) started to consolidate as important citrus producers as well [3]. The international fruit market moves annually around 20 billion US dollars, where Brazil participates with as little as US$104 millions.

In spite of its main role as a producer, Brazil is also an important fruit importer, with purchases reaching US$ 400 m. in 1996 [4].

The main difficulties for Brazilian exports of fruits are phytosanitary barriers. Just mentioning citrus, the main insect pests of economical importance in the State of Sao Paulo are: *Phyllocoptruta oleivora, Brevipalpus phoenicis,* the fruit flies *Ceratitis capitata* and *Anastrepha fraterculus.* We can also mention: *Xilella fastidiosa*, *Pinnaspis aspidistrae* and *Unaspi citri, Chrysomphalus ficus, Mytilococus beckii, Orthezia praelonga, Parlatoria cinerea,* among others [5]. Recently, the orange fruit borer, *Ecdytolopha aurantiana* which did not use to be an important pest for citrus, is now devastating some Brazilian orange farms because it is difficult to control [6].

There are numerous references in the literature about the efficiency of irradiation to control citrus pests [7][8][9][10]. Being so, there is a huge potential for the use of irradiation for quarantine purposes in the country, as soon as the technique is approved for importer countries. This is why detailed economic and financial feasibility studies for placing commercial food irradiation facilities are being considered.

In March 1998, the first 2,000 tons of Brazilian papayas were shipped to the USA as the clearance was obtained at the end of 1997. The papayas coming from two Brazilian producers from the North of the State of Espirito Santo have proven to be fruit flies free. In 1991, American imports of papayas were about US$ 3.6m. jumping to US$ 30m. in 1996, following a consuming increase of 50% per year. Also in 1996, the USA imported US$ 104m. of mangoes.

The demand for all kinds of fresh fruits is increasing from all types of consumers. Some markets ask for frozen packaged high quality products. Irradiated products can contribute to fulfill those requirements and also to overcome trade barriers.

1.3. Radiation sensitive indicators

For any kind of industrial irradiation, a dosimetry system must be applied in order to determine the absorbed doses that shall cover the absorbed dose range of interest and shall be calibrated before use [11]. Frequently, however, it is useful to also have a radiation sensitive indicator to visually determine whether or not a product has been irradiated, rather than to measure different absorbed dose levels. Indicators are used to show that a specific product has been exposed to ionizing radiation, but do not give a quantitative value of absorbed dose, and therefore are not a substitute for routine dosimeters used in routine process monitoring nor a complement to dosimetry.

As it is already established [1], exposure of fresh plant products subjected to infestation by insect eggs, larvae, pupae or adults to a dose of 300 Gy prevent the emergence of normal adult insects. In some cases, however, a dose of about 100 Gy is able to prevent the emergence of normal adults, when eggs or larvae are irradiated.

STERIN irradiation indicators are products of the International Specialty Products (ISP) of Wayne, New jersey, USA. These indicators were designed to provide visual verification of irradiation treatment at 50–500Gy dose levels and can be used as quality devices for irradiation disinfestation (required doses up to 1kGy). STERIN labels were designed as threshold indicators, where a visual message changes from “NOT IRRADIATED” before exposure to “IRRADIATED” after exposure at or above the threshold indication dose (e.g.125 Gy, 300 Gy).

The aim of this work was to evaluate STERIN label indicators to be used for Brazilian irradiated fresh fruits. As there are no industrial gamma irradiation facility in the country yet, able to deliver radiation doses recommended for quarantine purposes, e.g. below 1 kGy, the tests were performed using the radiation sources available at our institute, where most of the research on fruit irradiation is performed.

2. TESTS OF LABEL DOSE INDICATORS USING A GAMMACELL 220

ISP STERIN 125 and ISP STERIN 300 (International Specialty Products ISP Dosimeter Division of GAF Industries, Wayne, New Jersey, USA) were used. The manufacturer described that STERIN should be used as qualitative indicator since visual interpretation of the indicator opacity is only reproducible with an optical densitometer and not with a human eye. For that reason, both the visual changes in the indicators and spectrophotometric readings of the detached label indicator were evaluated.

The STERIN indicators were peeled and removed from the release sheet and attached on wooden supports. In experiment A, dose rate of about 428 Gy/h, doses of 5, 10, 50, 70, 125 and 200 Gy for STERIN 125 and 5, 10, 50, 200, 300 and 500 Gy for STERIN 300 were employed. In experiment B, another set of doses were used: 50, 100, 125, 150 and 200 Gy for STERIN 125 and 100, 200, 300, 400 and 500 Gy for STERIN 300, when the dose rate was 8.54 kGy/h. Tables 1 and 2 show the results of the irradiation of STERIN labels in a Co60 Gammacell 220 (AECL) for experiments A and B respectively. In all the assays, the indicator windows appeared completely dark, because the radiation sensitive film was fully opaque covering the word “NOT” displaying the visual message “IRRADIATED”, when the irradiation dose was 125 Gy and 300 Gy for STERIN 125 and STERIN 300 respectively. Nevertheless, they are not precise enough, as even smaller doses than the theoretical thresholds gave also the same indication.

After irradiation, the sensitive plastics were detached from the label and cleaned with ethyl acetate mixed with ricinus oil to remove the adhesive glue. Spectrophotometric measurements were performed using a Pharmacia LKB Novaspec II spectrophotometer. Thickness measurement was made with a Peacock micrometer and values of 0.573+/ 0.002mm for 125 Gy Indicator and 0.370+/ 0.002mm for 300 Gy Indicator were found.

Figures 1 and 2 show the absorbance at 665nm vs. dose curves for 125 Gy and 300 Gy Indicators. As can be seen, near the threshold for each one, a deviation from the linearity was observed.

TABLE 1. EXPERIMENT A, DOSE RATE 0.4 kGy/h. READINGS OF STERIN LABELS IRRADIATED IN A GAMMACELL 220. **()** **“NOT”** COULD BE SEEN CLEARLY; **(+)** **“NOT”** COULD HARDLY BE SEEN; **(++)** **“NOT”** WAS COMPLETELY COVERED

Dose (Gy)	**STERIN 125**		**STERIN 300**	
0				
5				
10				
50	+	+		
70	+	+		
125	++	++		
200	++	++	++	++
300	++	++	++	++
500			++	++

TABLE 2. EXPERIMENT B, DOSE RATE 8.5 kGy/h. READINGS OF STERIN LABELS IRRADIATED IN A GAMMACELL 220. **()** **“NOT”** COULD BE SEEN CLEARLY; **(+)** **“NOT”** COULD HARDLY BE SEEN; **(++)** **“NOT”** WAS COMPLETELY COVERED

Dose (Gy)	**STERIN 125**		**STERIN 300**	
0				
50	+	+		
100	++	++	+	+
125	++	++		
150	++	++		
200	++	++	++	++
300			++	++
400			++	++
500			++	++

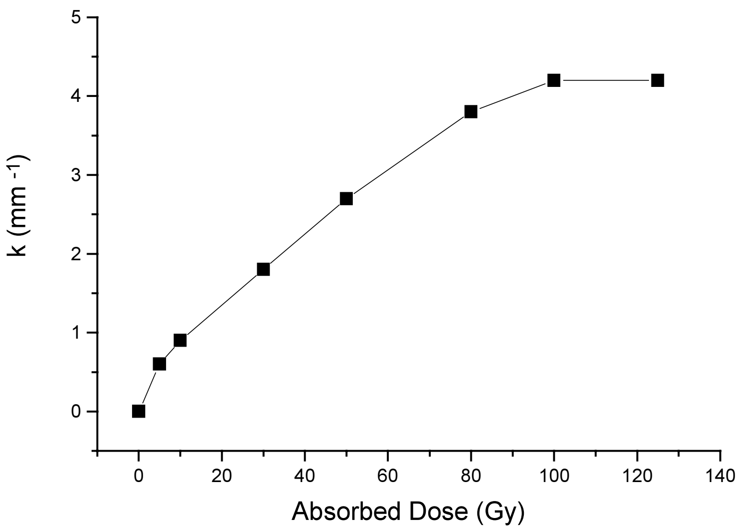

Figure 1. Dose response curve at 665 nm for 125 Gy INDICATOR irradiated with ^{60}Co gamma rays.

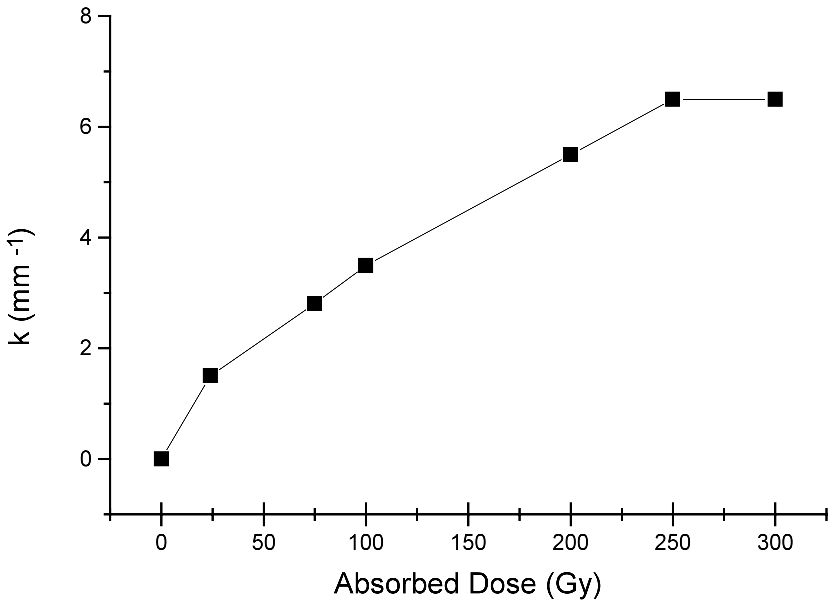

Figure 2. Dose response curve at 665 nm for 300 Gy INDICATOR irradiated with ^{60}Co gamma rays.

3. TESTS OF LABEL INDICATORS USING A PANORAMIC ^{60}Co SOURCE

Another kind of gamma source, a Panoramic Irradiator from YOSHISAWA KIKO Ltd. was also employed. The source itself is a pencil of ^{60}Co of 20 cm height and half an inch diameter. The dose distribution, that had been previously mapped by Fricke dosimetry, was checked with the STERIN indicators. Table 3 shows the readings of the two kinds of indicator labels which were irradiated onto wooden supports of 20 cm in height, previously distributed on the table of the irradiation camera. The irradiation doses were: 0, 61, 84, 124 and 197 Gy for the STERIN 25, and 0, 124, 197, 354 and 656 Gy for the STERIN 300 indicator. Similarly as before, the radiation sensitive film was fully opaque covering the word "NOT" displaying the visual message "IRRADIATED", when the irradiation dose was 125 Gy and 300 Gy for STERIN 125 and STERIN 300 respectively. Once the minimum radiation indicators proved to work, it was decided to use them on a normal wooden package for papaya or orange fruits. As a phantom for the fruits, latex balloons filled with water were used. In this case, samples of STERIN were employed distributed all over and into a wooden box, as can be seen in Fig. 3. In this case, a minimum dose of 200 Gy, at the farthest point from the source, was applied. Slight differences in the opacity of the labels were found at different points of the box. Nevertheless, all the readings can be considered as "IRRADIATED".

Some experiments were made with the STERIN labels attached in front of the fruit packed in plastic nets (containing 3 to 4 pieces) or behind the fruit on wooden supports. In this experiment, three kinds of citrus were used: two varieties of oranges and soft citrus, irradiated with 300, 139 and 114 Gy, with the labels placed in front and behind the fruits (Table 4).

TABLE 3. READINGS OF STERIN LABELS IRRADIATED IN A PANORAMIC GAMMA SOURCE. **()** **"NOT"** COULD BE SEEN CLEARLY; **(+)** **"NOT"** COULD HARDLY BE SEEN; **(++)** **"NOT"** WAS COVERED COMPLETELY

Dose (Gy)	**STERIN 125**		**STERIN 300**	
0				
61	+	+		
84	++	++		
124	++	++	+	+
197	++	++	++	++
354			++	++
656			++	++

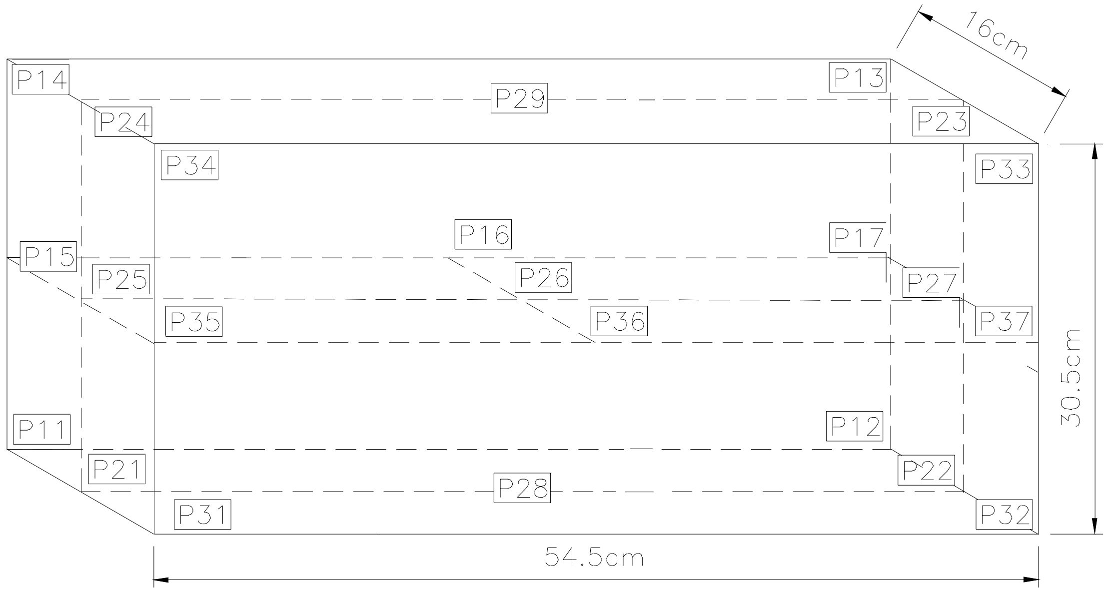

FIG. 3. Distribution of label dose indicators in wooden boxes.

TABLE 4. READINGS OF STERIN LABELS ATTACHED TO SOME FRUITS IRRADIATED IN A PANORAMIC GAMMA SOURCE. **()** **"NOT"** COULD BE SEEN CLEARLY; **(+)** **"NOT"** COULD HARDLY BE SEEN; **(++)** **"NOT"** WAS COMPLETELY COVERED

Dose (Gy)	**STERIN 300 in front**		**STERIN 300 behind**	
0				
114	+	+	+	+
139	+	+	+	+
<300	++	++	++	++
>300	++	++	++	++

4. TESTS OF LABEL DOSE INDICATOR USING AN ELECTRON BEAM ACCELERATOR

The label indicators were also assayed for irradiation with an electron beam (EB) accelerator, a Dynamitron, Radiation Dynamics Inc., 1.5 MeV. Calorimetry is the method used for dosimetry in EB irradiation. Due to limitations of the machine, the minimum doses that were possible to be attained were about 268 Gy (0.3 mA), 357 Gy (0.4 mA), 446 Gy (0.5 mA) and 536 Gy (0.6 mA), depending on the current. In this case, all the label readings can be considered equally as “IRRADIATED”.

5. OTHER DOSIMETERS

Frequently for fruit irradiation, Gammachrome YR from Harwell Laboratory dosimeters are used (dose range 0.1 to 3 kGy) at IPEN. Calibration data are generated at Harwell and supplied to users as examples or for comparing with users data. Our calibration curves are prepared whenever a new lot of dosimeters is used. Figs. 4 and 5 present the calibration curve of Gammachrome YR obtained from dosimeters irradiated in the Gammacell 220 and a panoramic source for the dose range 0.11 kGy. Fig. 6 presents the dose response curves of another dosimeter, DM1260 films (Far West Technology, CA) from 0.02 to 3kGy, read at 510 nm, where on the ordinate axis k values are plotted, being $k = (AA_0)/x$, where A stands for the absorbance of the irradiated dosimeter, A_0 is the absorbance of the unirradiated dosimeter and x the thickness of the dosimeter.

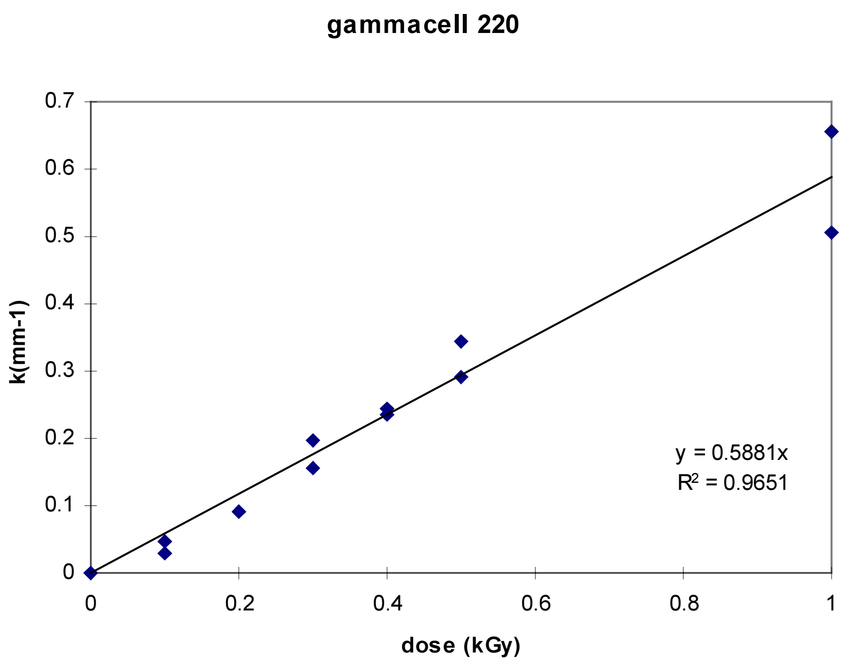

FIG. 4. Specific absorbance (530 nm) versus dose, Gammachrome YR, batch 5, Gammacell 220.

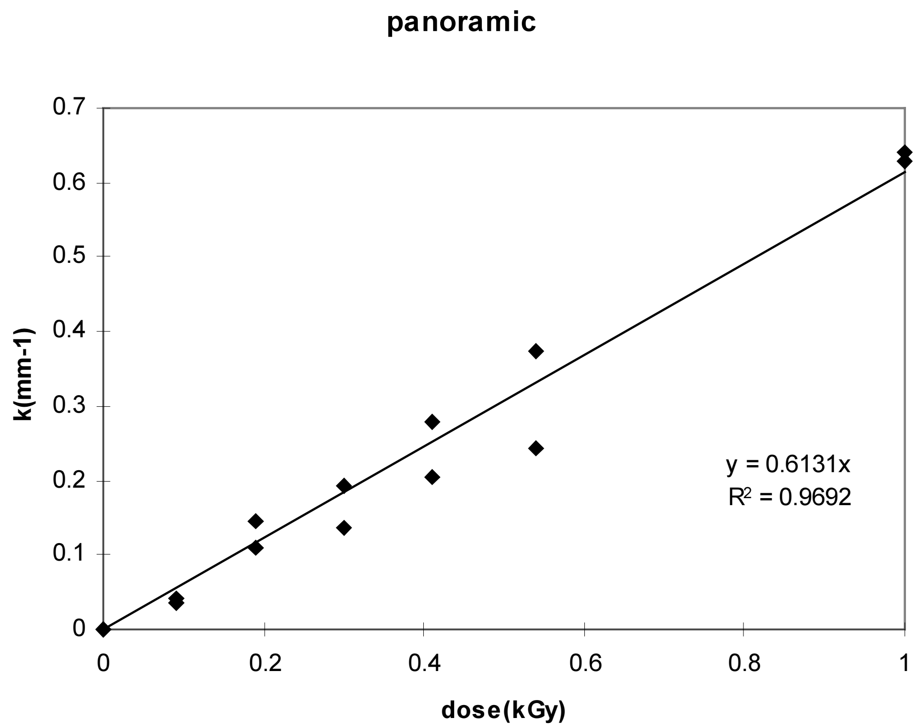

FIG. 5. Specific absorbance (530 nm) versus dose, Gammachrome YR, batch 5, panoramic ^{60}Co source.

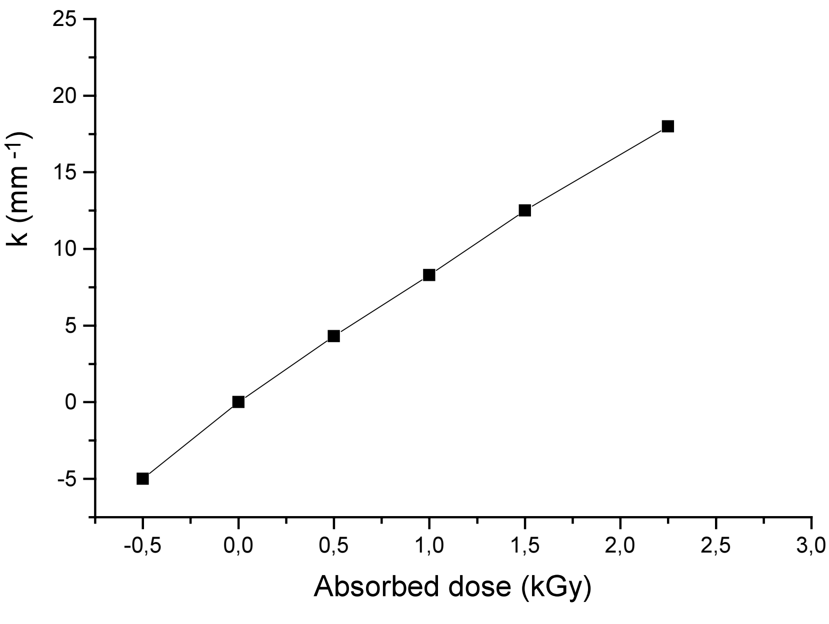

FIG. 6. Dose response curve for DM1260 films (spectrophotometric readings at 510 nm).

6. FINAL REMARKS

Our country is going to become an important exporter of fresh fruits such as oranges, mangoes, papayas, melons, apples, bananas, pineapples, guavas, limes, passion fruits, avocados, figs, plums, peaches, strawberries, carambolas and Barbados cherries. Dried fruits and nut trees are also important items of internal and external trade. Brazil, together with Argentina, Uruguay and Paraguay is now working on the feasibility of MERCOSUR. The agreement on Sanitary and Phytosanitary Measures to be implemented with the establishment of the World Trade Organization concerns the application of food safety as well as animal and plant health regulations. Food irradiation will surely be part of its future discussions. In that context, the measurement of gamma radiation quantities, e.g., absorbed dose in materials such as plastics or on foodstuffs itselves is a convenient means of quality assurance in radiation processing.

The STERIN label dose indicators can be useful tools provide they can be produced inexpensively and in large quantities making suitable for food radiation processing.

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REFERENCES

[1] ICGFI. Code of Good Irradiation Practice for Insect Disinfestation of Fresh Fruits, ICGFI Document N^o 7, Vienna (1991).

[2] SEATON, L. (Ed.), Fresh Produce Desk Book, Published by Fresh Produce, London (1998) 34.

[3] ANOM. Panorama da Citricultura Brasileira. Circuito Agícola, ABRACEN, No. 37, Sao Paulo (1996) 79.

[4]. FRANCO, L., KASSAI, L., Maior produtor exporta pouco. In: Agribusiness. A Gazeta Mercantil, Sao Paulo, 25 December (1997).

[5] DE NEGRI, J.D., Cultura dos Citrus. Boletim Técnico CATI No. 228, Campinas, Sept. (1996).

[6] FARIA, J.T., Use of Radiation as a Quarentenary Process to Control the orange Fruit Borer Ecdytolopha arantiana (Lima, 1927) (Lepidoptera, Tortricidae) on orange Fruits (Citrus sinensis), Variety "Pera" and Observations on its Effects on Fruits Quality. Ph.D. Thesis, IPENUniversity of Sao Paulo, 29 August 1997, Sao Paulo, Brazil.

[7] BURDITT Jr., A.K., et al., Lowdose irradiation as a treatment for grapefruit and mangoes infested with Caribbean fruit fly larvae. Washington, USDA/Agriculture Research Service, 1981. 9p.

[8] MONTALBAN, A., et al., Irradiacion para el comercio internacional de productos agropecuarios. El caso de los citricos. In: Cost–benefit Aspects of Food Irradiation Processing Symposium, Aix en Provence, 1993, Proceedings, Vienna, IAEA (1993) 321–328.

[9] LIU, Z., et al., The Study on Preservation of Nanfeng Mandarin by Radiation. Journal of Radiation Research and Radiation Processing **6(**30): 48–52, 1988.

[10] OMAHONY, M.A.D.P., GOLDSTEIN, L.R. Methods for sensory evaluation of Navel Oranges treated with Electron Beam Irradiation. Lebensmittel Wissenschaft und Technologie **20**(2):78–82, 1987.

[11] O'HARA, K., THAYER, D., METHA, K., Standard E10.01Z Guide for Dosimetry in Radiation Research on Food and Agricultural Products Annual Book of ASTM Standards, Washington (1996).

DEVELOPMENT OF STANDARDIZED METHODS TO VERIFY ABSORBED DOSE OF IRRADIATED FRESH AND DRIED FRUITS, TREE NUTS IN TRADE

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Abstract. Investigations were carried out on standardization of desired process control parameters such as dose distribution in trade containers, container standardization and development of "label" dosimeters. A prototype "label" dose indicators Sterins for threshold doses of 125 Gy and 300 Gy was studied. Dose distribution was studied using fresh fruits and tree nuts in trade and standardized containers with varying product densities. The distribution of absorbed doses was measured by Fricke, Gammachrome YR, clear Polymethylmethacrylate (PMMA), EthanolChlorobenzene (ECB) and Sterin 300. These values are given as Dmax/Dmin ratios in relation to product bulk densities. It was observed that bulk densities varied greatly among different products depending on the types of fruits, containers and pattern of loading which also affected dose distribution. Dmax/Dmin obtained by proper dose mapping could be kept low by arranging proper irradiation conditions which ensured uniform dose distribution. Prototype "label" dose indicators like Sterins and clear PMMA were used for dose mapping along with the standard primary and secondary dosimeters. Sterins and clear PMMA were also studied for their dosimetric properties, particularly for use in label dosimetry. Sterins 125 and 300 evaluated visually showed their integrity at their threshold doses. The word NOT on Sterin 125 eclipsed after 115 Gy and on Sterin 300 after 270 Gy dose. Clear PMMA samples of 410 mm thickness irradiated at 200–1000 Gy showed linear response and had postirradiation stability for over a month storage at normal temperatures (21–35^{0}C) and humidities. These could be investigated further for developing as "label" dosimeters in insect control quarantine treatment. Other low dose indicators studied such as coloured perspex, dye solutions were not found useful at quarantine dose levels. Further investigations are required for developing a "label" dosimeter for commercial use.

1. INTRODUCTION

Fresh and dried fruits and tree nuts are often infested by a variety of insect pest species, many of which result in the products coming under quarantine restrictions by the importing countries. With the continuing tightening of restrictions on, and outright banning of the fumigants for insect pest control, there is increasing interest on the part of the regulatory authorities and affected agroindustries in low dose irradiation treatment as a proven nonchemical alternative treatment method. Low dose irradiation (0.15–1.0 kGy) does not always result in insect death but leads to sexual sterilization plus failure to develop from eggs/larvae to normal functioning adults [1–3]. Successful application of this method depends greatly on the delivery of the required overall absorbed dose, particularly the minimum effective dose which is to be ascertained and measured by suitable dosimeters. This assessment of the treatment is important to the inspectors for ensuring control of insects. This assessment is visualized to be done by a quantitative "label" dosimeter that can be affixed to the outsides of the product cartons and read by inspectors using a hand-held scanner. Both need to be developed and it is a challenging approach.

To meet the challenge the CRP on Standardized Methods to Verify Absorbed Dose in Irradiated Fresh and Dried Fruits, Tree Nuts in Trade was initiated by International Atomic Energy Agency in December, 1993, participated by research contract holders from several member states with the intended objectives:

1) to develop a quantitative " label" dosimeter and hand-held or otherwise a simple onthespot reader, and
2) to standardize its applications to such an extent that the dose is read accurately and reliably relatable to the minimum dose received in the container of the product which may harbour live insects against which irradiation is targeted to achieve quarantine security/commodity disinfestation.

Aiming at these objectives the research work carried out during the last 4 years has been reported in this paper.

2. MATERIAL AND METHODS

2.1. Procurement of experimental samples

Fresh fruits such as mangoes, guavas, pineapples, bananas, papayas, oranges and tree nuts were collected from gardens/traders. Paper cartons and wooden crates of 42×32×32 cm and 47×31×27 cm (LXBXH) sizes respectively, were prepared for dose delivery of the fruits and nuts. PMMA samples obtained from local markets and from Pakistan were used for this experiment. Coloured plastic samples obtained locally were also tried. Sterins were supplied by the International Specialty Products, New Jersey, U.S.A.

2.2. Measurement of product bulk densities

Fruits and nuts were packaged in commercial containers. Bulk weight was recorded and container volume was measured for getting the bulk densities. The variations in bulk densities with the kinds of fruits, containers and packaging pattern were also observed.

2.3. Dose distribution measurement

The fruits in the containers with different bulk densities were irradiated in a Co60 gamma source (Gamma beam 650, AECL, Canada). The irradiator was a dry, pneumatic type and the arrangement of the source was panoramic. The containers were placed in different standardized positions which were calibrated by Fricke dosimeters. The doses absorbed by the fruits were measured by Fricke, Gammachrome YR, amber perspex and ethanolchlorobenzene dosimeters placed in different positions of the container and the dose received by each dosimeter was read. For uniform distribution of the dose the containers were turned manually 4 or 2 times during irradiation.

2.4. Development/evaluation of the dosimeters

2.4.1. Evaluation of Sterin 125 and Sterin 300

Sterin is a coloured dose indicator which completes its colour change at defined threshold dose. Sterins were attached to different positions in the fruit containers having different bulk densities and irradiated, turning the containers once for adequate dose distribution. The doses applied were around 30–40 percent above and below the threshold to facilitate visual estimation.

2.4.2. Development of clear PMMA dosimeters

Clear PMMA (polymethylmethacrylate) of local and Pakistan origin were used for this study. Common grade PMMA samples were of 4, 5 and 6 mm thickness and the special grade of 4, 6, 8 and 10 mm thickness. These were cut into standard cuvette sizes, washed in spirit, dried and packaged individually (not sealed) in aluminium foils. Absorption spectra of these samples was determined and peaks were recorded. Two sets of experiment were carried out one set was irradiated to 200–1200 Gy at 200 Gy dose intervals and the other at 200–800 Gy at 100 Gy incremental doses, both in triplicate at ambient aerobic condition.

Irradiated local and Pakistani PMMA were read at 300 nm and 305 nm respectively.

2.4.3. Development of coloured low dose indicators

A coloured low dose indicator solution was prepared using ferrous ammonium sulphate, benzoic acid, sulphuric acid and xylenol orange dye The dosimetric solution contained 0.20 mM ferrous ammonium sulphate, 5.0 mM benzoic acid and 0.20 mM xylenol orange in 0.05 N sulphuric acid. This is a yellow coloured solution which changes slowly at room temperature storage. The colour is stable under chilling condition. After irradiation, the violet colour produced is very stable even at room temperature [4]. The stability of the colour before and after irradiation was tested both visually and spectrophotometrically. This indicator was tested for colour change both visually and spectrophotometrically in response to irradiation doses 50–200 Gy.

Locally prepared coloured plastics and strips samples were tested for their dosimetric properties by irradiating at low doses. Four types of such coloured plastic items have been developed and absorption spectra studied. These were green, amber, red and blue coloured with absorption maxima of 430, 478, 518 and 602 nm respectively.

2.4.4. Postirradiation stability of the dosimeters

Irradiated Sterin, clear PMMA, coloured low dose indicator and Gammachrome YR were analyzed/observed for about 16 months for studying postirradiation stability of the dosimetric properties. Sterin was observed for 6 months, Gammachrome YR and clear PMMA for 1 month; all at ambient temperatures and humidities.

3. RESULTS AND DISCUSSIONS

Insect infestation is a problem in international fruit trade. The plant protection authority wants a simple and speedy detection method for ensuring observance of treatment for control of insects in the imported fruits and nuts. In case of irradiation disinfestation treatment a "label" indicating the efficacy of the treatment is wanted by the regulatory agency. This "label" dosimeter will indicate the absorbed dose from which the minimum absorbed dose could be derived by relating to product bulk densities and process operation.

3.1. Product bulk densities of fruits and dose distribution

Table 1 shows the bulk densities of different fruits packaged in different containers used in trade. Bulk densities were found to vary with the types of fruits, containers and packaging arrangements. Compact packing yielded higher density. Tree nut had high bulk density because of compact packing in containers (0.58–0.72 g/cc).

TABLE 1. BULK DENSITIES OF FRESH FRUITS IN TRADE CONTAINERS

Fruits	Bulk densities (g/cc)	
	Paper cartons	Wooden crates
Mangoes	0.44 – 0.50	0.54 – 0.58
Bananas	0.45 – 0.52	0.50 – 0.56
Pineapples	0.49 – 0.55	0.56 – 0.58
Guava	0.56 – 0.58	0.56 – 0.63
Papayas	0.44 – 0.47	0.44 – 0.48
Nuts	0.58	0.72

Container specification: L×B×H in cm
Paper Cartons: 42×32×32
Wooden Crates: 47×31×27

Data on dose uniformity ratios and overall average absorbed doses with different bulk densities of fruits are shown in Table 2. The dose distribution was measured by Fricke, Gammachrome YR, clear PMMA and ethanolchlorobenzene [5–7] to check their comparative performance. It is seen from the data that the dose uniformity values showed in general an increasing trend with increase in the bulk densities. The lowest dose uniformity ratio of 1.27 was recorded in the case of product bulk density of 0.45 g/cc and the highest of 1.50 at bulk density of 0.56 g/cc as measured by Fricke. Gammachrome YR, clear PMMA and ECB dosimeters were found to yield slightly higher values compared to the Fricke (Table 2). The dose uniformity ratios obtained in irradiation operation reflect dose distribution and for achieving better product quality, efficacy and economics it is desirable to keep the ratios as low as possible which can be achieved by selecting relevant materials and methods including product containers.

TABLE 2. Dmax/Dmin RATIOS AND OVERALL AVERAGE ABSORBED DOSES AT DIFFERENT BULK DENSITIES OF FRUITS AS MEASURED BY DIFFERENT DOSIMETRY SYSTEMS

Product bulk densities (g/cc)	**Dmax/Dmin ratios**: overall average absorbed doses in kGy by different dosimeters			
	Fricke	Gammachrome YR	Clear PMMA	ECB
0.04	**1.16** ;			
0.45	**1.35** ; 0.20			
0.49	**1.40** ; 0.21		**1.50** ; 0.25	**1.64** ; 4.37
0.56	**1.50** ; 0.22	**1.56** ; 0.41	**1.64** ; 0.38	**1.88** ; 2.84
0.66	**1.47** ; 0.24	**1.71** ; 0.52	**2.00** ; 0.39	**1.94** ; 2.44

3.2. Sterins

Sterins are minimum radiation dose “ Indicators”. Two types of Sterins have been supplied one completes its colour change at 125 Gy and the other at 300 Gy. When Sterin 125 exhibits a complete colour change the indicator received at least 125 Gy; complete colour change of Sterin 300 indicates an absorbed dose of 300 Gy received by the indicator.

TABLE 3. VISUAL ESTIMATION OF COLOUR CHANGE OF STERIN 300 SUBJECTED TO PREPOST THRESHOLD IRRADIATION DOSES

Dosimeter positions	Dose measured by Fricke (Gy)	Eye estimation of colour changes
1	182	Light darkening ; NOT visible
2	270	Getting darkened ; NOT blurred
3	311	Total darkening ; NOT invisible
4	353	,, ,, ; ,, ,,
5	326	,, ,, ; ,, ,,
6	291	Darkened ; NOT invisible
7	215	Light darkening ; NOT visible
8	245	Darkening ; NOT indistinct
9	305	Total darkening ; NOT invisible
10	320	Total darkening ; NOT invisible.

Sterin 125 dosimeters placed in the product was found to complete its colour change at 125 Gy in a set of experiment carried out at 70–156 Gy dose range. Visually the indicator began showing its effectiveness at 118 Gy the word “NOT” becoming hardly readable leaving the message “IRRADIATED” in the label for certification of the product. Sterin 125 dosimeters subjected to 50–200 Gy dose range performed as expected showing the peak at 125 Gy of irradiation dose which was observed by O.D. measurement read at 550 nm. Sterin 125 may find commercial application for fruit fly control. The minimum dose for quarantine treatment in this case is expected to be 125 Gy.

Sterins 300 subjected to Dmax 335 Gy and Dmin 269 Gy was found to complete its colour change at 297 Gy, close to the threshold dose of 300 Gy. These were irradiated in trade container, tomatoes and guavas serving as phantom products. Visual estimation of the colour change of Sterin 300 at 10 percent ± threshold dose also confirmed the integrity of the indicator. The word “NOT” could be read up to 260 Gy under illumination but after that it became hardly readable. At and above the threshold doses colour change became progressively deeper (Table 3). Other work also confirmed these results. They found that the Sterin labels appeared adequate not only for visual identification but also for obtaining rough dosimetric data. This was further supported by optical density vs dose study for Sterin in which the curve showed the peak at 300 Gy (Fig. 1).

Sterin 300 may find application as a label dosimeter in insect pest quarantine control treatment. The indicator signals seem to remain relatively stable with time and within simulated trade and environmental condition for fresh and dried fruits and tree nuts.

3.3. Clear PMMA

Clear PMMA of 4, 5, 6, 8 and 10 mm thickness was investigated for dosimetric properties and possible use as a label dosimeter in quarantine control treatments. Two sets of experiments were conducted one with Pakistani PMMA of 4, 6, 8 and 10 mm thickness and the other with local PMMA of 4, 5 and 6 mm thickness. Both sets were irradiated to 200–1200 Gy at 200 Gy dose intervals. The results are shown in Figs 2 and 3. It is seen from the Fig. 2 that the specific absorption in relation to the absorbed dose yielded linear curves in case of 4, 6, 8 and 10 mm PMMA. The results were fairly reproducible but slight deviations were noted from sample to sample. The PMMA samples also gave significantly measurable response at low incremental doses of 50/100 Gy. This property may qualify it for detecting dose variation of 50 Gy. This is significant for label dosimetry; but for practical use the dosimeter should indicate dose difference close to 10 percent from the target dose.

Local PMMA samples showed lower absorption and slightly inconsistent results compared to the special grade (Fig. 3) at 200–1200 Gy irradiation at 200 Gy intervals. The curve was almost linear up to 800 Gy but deviation occured after 1000 Gy. These samples were less homogenous and it might be one of the reasons for this inconsistent behaviour.

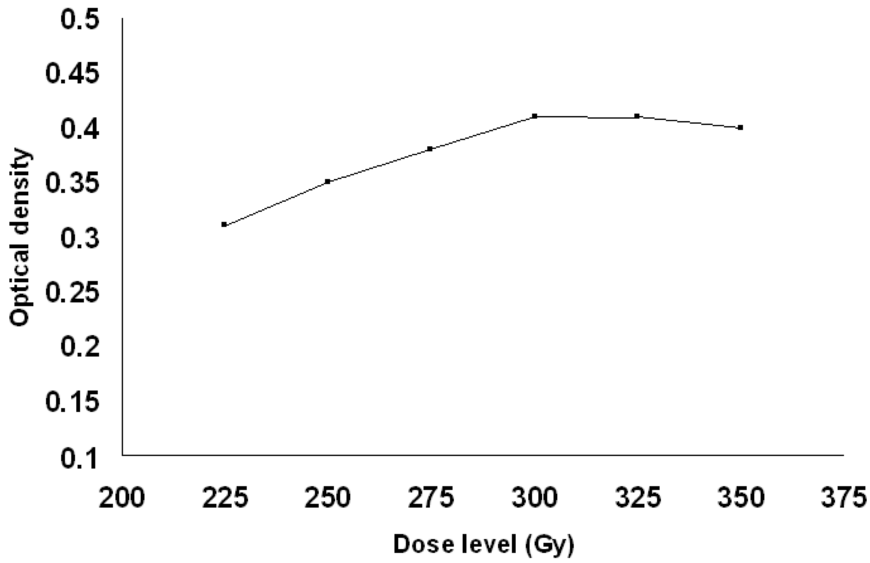

FIG. 1. Optical density vs dose for sterin 300.

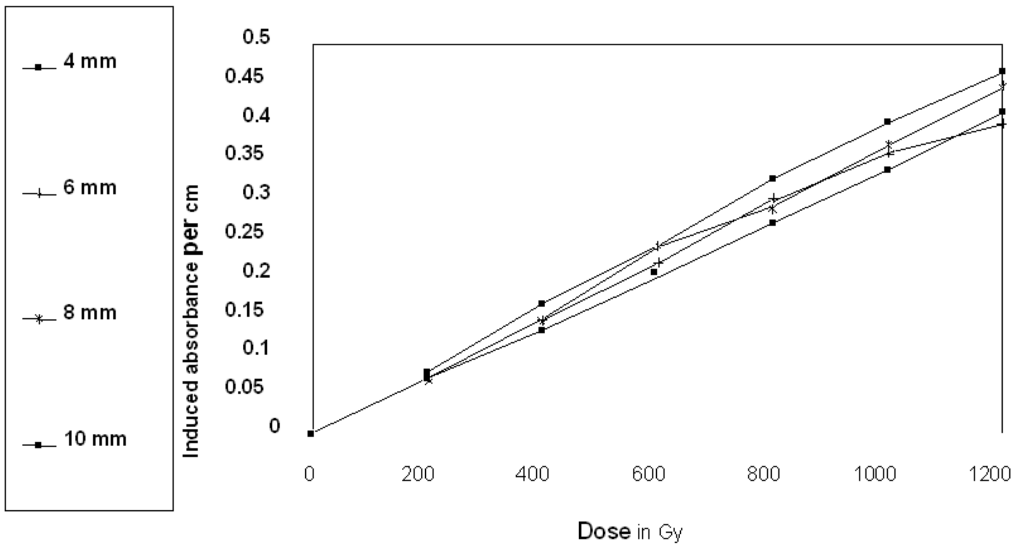

FIG. 2. Dose response of Pakistani clear PMMA.

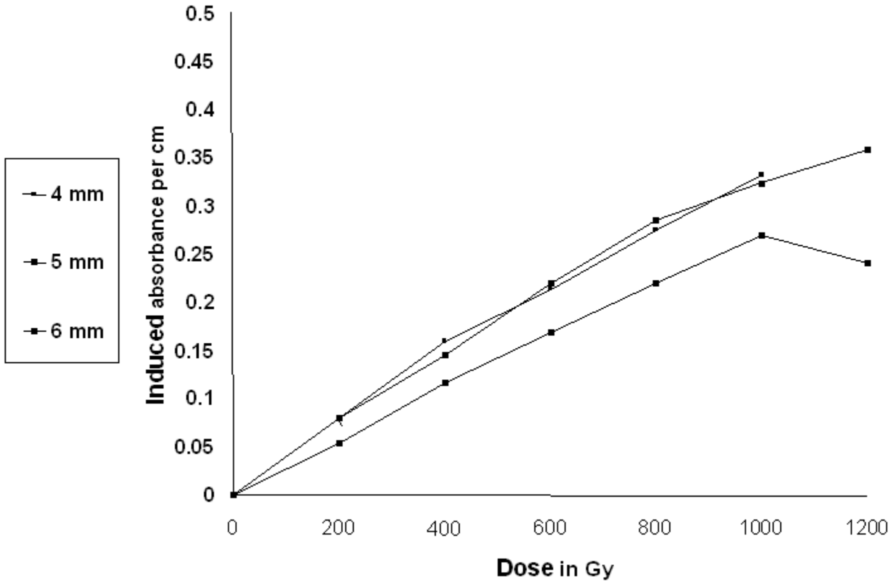

FIG. 3. Dose response of local clear PMMA.

3.4. Coloured low dose indicators/dosimeters

The xylenol dye solution containing ferrous ammonium sulfate, benzoic acid and sulphuric acid was yellow in colour and completed its colour change to violet at 100 Gy of irradiation dose. The intensity of colour increased with dose. This indicator provides a convenient tool to visibly assess if the product is irradiated (above 100 Gy).

The dosimetric property of locally developed coloured plastic was investigated. Irradiation of these materials at 200–2000 Gy dose showed low and nonmeasurable response below 1500 Gy of gamma irradiation.

3.5. Post-irradiation stability of dosimeters

Sterin 125 or 300 kept below 10^0C for 6 months and then irradiated were found to perform as expected. Irradiated Sterins stored at 20–35^0 C and 40–100 percent humidity for about 9 months retained their integrity. Those dosimeters received lower than the threshold dose displayed the word "NOT" and those at or above the threshold levels the word "IRRADIATED"; "NOT" was invisible.

Both local and Pakistani grade PMMA studied were found to retain their dosimetric property 1 month or more after irradiation and storage at ambient temperatures (22–35^0C) and humidities (40–90% RH) (Fig. 4). The dosimeters showed slight increase in the absorbed dose value during 1st week of irradiation and became stable thereafter.

3.6. Application as "label" dosimeters

Sterin 125 may be used in fruit fly quarantine treatment. Sterin 300 can be used in treatments requiring minimum 300 Gy absorbed dose received by the product. This can be ensured by adopting appropriate process control.

Pakistani PMMA of 610 mm thickness may be further investigated for use as "label" in treatments requiring 0.3 to 1 kGy absorbed doses. The signal seemed relatively stable and dose uniformity ratios were comparable (Table 2). Coloured low dose indicators like Sterins could be developed by using combination of dyes for carrying the message as in the sterins.

Gammachrome YR may be considered for use as a label dosimeter with two limitations: 1) it may not be useful below 300 Gy, and 2) at that dose level it fades about 50 percent in two weeks postirradiation storage.

A hand-held scanner needs to be developed for rapid screening of the " label" dosimeter at the entry points.

4. CONCLUSION

A "label" dose indicator is needed for promoting international trade of irradiated fruits and nuts. This is required for indicating the efficacy of the insect control in irradiation treatment. A prototype label, Sterins showed integrity at the irradiated absorbed doses. Clear PMMA and low dose indicators studied showed promise as absorbed dose indicators, but needs further investigations for use as "label" dosimeters. Proper and defined process control procedures are required for successful dose monitoring by the "label" which in turn may make the job of inspection easy, speedy and accurate.

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REFERENCES

[1] BHUIYA, A.D., M. AHMED, G. NAHAR, Radiation Disinfestation of Pulses, Oilseeds and Tobacco leaves. IAEARc273.3/2, Insect Disinfestation of Food and Agricultural Products by Irradiation (1991), IAEA, Vienna.

[2] MATIN, A. S. M. ABDUL, Susceptibility of Rice Weevil to Gamma irradiation. Internal paper, AECD/AG/15 (1966), AECD, Dhaka.

[3] PESSON, P., Some experimental Data on Co60 radiation dose capable of arresting insect infestation of cereals and flour. Food Irradiation **3(4)** A18 21 (1963).

[4] BHAT, R.M., G.R. NARAYAN, B.L. GUPTA, Bulletin of Radiation Protection, **10** (122) 21 (1987).

[5] AMERICAN SOCIETY FOR TESTING AND MATERIALS, Guide for selection and application of dosimetry systems for radiation processing of food. ASTM Standard E1261 and ASTM Standard E1276 on PMMA and radiochromic film dosimeters (1988).

[6] WHITTAKER, B. The Gammachrome YR system. UK Gamma panel symposium, Dosimetry and Control of Radiation Processing, National Physical Lab., UK (1987).

[7] KOVACS, A.; V. STENGER; G. FOLDIAK and L. LESEZE., Evaluation of Ethanol monochlorobenzene dosimeters by the Conductivity method. IAEA SM272/33. Proc. Intl. Symp. On High Dose Dosimetry, Vienna, October (1984).

[8] EHLERMANN, D.A.E. Label dosimetry as a means to verify dose. Report presented at FAO/IAEA 2nd RCM on Standardized Methods to Verify Absorbed Dose in Irradiated Fresh and Dried Fruits, Tree Nuts in Trade, Karlsruhe, Germany, September (1996).

DEVELOPMENT OF LABEL DOSIMETERS AND ANALYTICAL METHODS TO VERIFY ABSORBED DOSE IN IRRADIATED DRIED FRUITS/TREE NUTS

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Abstract. Density measurements of fresh/dried fruits and tree nuts varied depending upon the package size, type and the nature of the sample. For the development of label dosimeters, the samples of clear PMMA, in the thickness range of 410 mm gave a linear response in relation to the irradiation doses (0.125–1.0 kGy) and the optical response was stable almost for 6 months at ambient storage (20–35°C; R.H 40–80%). Flexible polymers (polyethylene & PVC) materials were not found suitable in the dose range of 0.1–3.0 kGy. Subjective evaluation of Sterin indicator, an ISP product from USA, revealed that this new material is generally reliable, however, they were also affected by the doses lower than their threshold values (125 and 300 Gy) as well as exposure to light during storage. The yellow PMMA dosimeter (YLPMMA) developed by China was useful in the range of 125–1000 Gy of gamma radiation. Dose distribution studies of research irradiator at NIFA and a commercial gamma source (PARAS) at Lahore, indicated almost a good dose uniformity in the product containers in each case. Among the analytical methods (thermoluminescence, chemiluminescence and gas chromatography) the thermoluminescence measurements exhibited clearly reproducible and dose dependent differences between treated (0.5–1.5 kGy) and untreated samples of dried fruits/nuts.

1. INTRODUCTION

Irradiation processing of foods extends shelf-life, minimises food losses, enhances the quality of fresh produce and can disinfect stored products. It can also be used as quarantine treatment, and improves the hygienic and microbiological quality of different food materials. In about 40 countries, more than 50 different kinds of foods are now approved for irradiation treatment. The work conducted by FAO/IAEA through coordinated research programmes (CRPs) has shown conclusively that there is no adverse effect from consumption of foods irradiated up to 10 kGy [1]. Due to differences in national regulations concerning food irradiation, it is important to ensure the proper control of irradiated foods in international trade. The existence of different requirements and standards in different countries constitutes a barrier to international trade. Different analytical techniques have been applied [25] to detect whether a particular food has been irradiated or not. In view of global interest in food irradiation processing, there is therefore, a need for label dosimeters and detection methods for irradiated food. In this context a final RCM of FAO/IAEA research coordinating programme on analytical detection methods for irradiation treatment of foods (ADMIT) was held in Belfast, UK 2024 June, 1994. A new CRP of FAO/IAEA on standardised methods to verify absorbed dose in irradiated fresh and dried fruits and tree nuts in trade has been completed in April 1998.

Stick-on labels are also a possible form of radiation monitoring devices to be used in the control of food irradiation. Two potentially important systems have been released, i.e. GafChromic Dosimetry Media dosimeter film and Sterin indicator labels (ISP, NJ. USA). Recently usefulness of Sterinindicator has been reported [6, 7]. Where the evaluation of this label dosimeter is further needed, the information as to whether or not the minimum dose effective for the intended treatment has been applied throughout the treated sample, remains to be resolved. Research findings of the Food Science Group at NIFA concerning the work on development of analytical methods and label dosimeters for irradiated foods have been reported [8–15]. Recently Government of Pakistan has finally issued a gazette notification approving the treatment of food by gamma irradiation [16], and hence a case for installation of food irradiator along with the already operational commercial irradiator for medical supplies (PARAS) at Lahore, becomes strong. In view of the potential of food irradiation processing in this country, the present studies were conducted to develop reliable analytical methods and/or label dosimeters for measuring absorbed gamma irradiation doses in fresh/dried fruits and tree nuts. The present report deals with the results obtained in these studies.

2. MATERIALS AND METHODS

The samples of dried fruits (apricot, date, raisin), fresh fruits (apple, mango) and tree nuts (almond, pinenut, walnut) were obtained in fresh condition from the wholesale market at Peshawar. In order to develop some suitable dosimeters, the indigenous samples of polymethylmethacrylate (PMMA) were provided by the commercial manufacturer (PakPoly. Industries (Pvt) Ltd Karachi), which were protected by the stickon paper to avoid any scratches during irradiation and storage. The stickon papers were removed before spectral measurements. The flexible polymers (polyethylene films) were obtained from the Packages Ltd, Lahore. The samples of dried fruits and tree nuts as well as fresh apples and mangoes were packed in polyethylene pouches for radiation treatment and subsequent detection tests.

2.1. Density measurement

For fresh fruits, wooden boxes of different sizes are used for holding and inland transportation in Pakistan. In the case of dried fruits and tree nuts, the material is held or used for inland trading in the nylon/jute bags with capacity of 520 kg. Paper cartons or hardboard boxes are also used as storage containers for holding or short distance transportation. Wooden boxes have the advantage that they can be reused and therefore densities of food materials in the standardised wooden container (40 × 30 × 20 cm) and nylon bags (10 kg) were measured. The density of fresh fruits, dried fruits and tree nuts was also measured in trade containers used for overseas transportation such as hardboard cartons of different sizes depending upon the nature of the material. The most common sizes of cartons used in the country are 40 × 35 × 30 cm and 45 × 40 × 35 cm for fresh and dried fruits respectively.

2.2. Irradiation

The dried fruits and tree nuts samples were irradiated at doses of 0.5, 1.0 and 1.5 kGy and the fresh apples and mangoes treated with radiation doses of 0.2, 0.4, 0.6, 0.8 and 1.0 kGy. In the case of label dosimeters, the PMMA and polyethylene samples were irradiated with a dose range of 0.1–3.0 kGy. Irradiation of the materials was carried out in a Co60 source ISSLEDOVATEL (CIS) at a dose rate ranging 2.41–2.46 kGy/hr (over 4 year period) with max/min ratios 1.05–1.07. Dose distribution of the source was determined by Fricke dosimetry [17].

2.3. Dose distribution

Dose distributions of Co60 gamma ray research irradiator (ISSELDOVATEL, CIS) at NIFA Peshawar, and Pakistan Radiation Services (PARAS) commercial irradiator installed at Lahore, were measured using 2 mm red polymer PMMA manufactured by the Pak. Poly Industry (PPI), Karachi under the trade name of PLASTIGLASS.

Research unit at NIFA consists of cylindrical radiation chamber having inside dimension of 15 cm diameter and 24 cm height. The source cage is made up of stainless steel frame, which holds up doubly sealed Co60 capsules. The capsules are arranged in a vertical position having free space available for loading of additional source pencils. The Co60 pencils are positioned in annular symmetry around the cylindrical radiation chamber. For depth dose measurements of this research iradiator, PMMA sheets of red colour (2 mm) in appropriate size were positioned vertically and horizontally in different planes in loaded and unloaded container. After irradiation of the sheets with dose of 2.5 kGy, they were cut in to small strips and absorbance measured at 640 nm using UVVis. Shimadzu Model 160 spectrophotometer and the isodose curves drawn. The isodose curves were normalized with respect to the dose at the central region, taken as 100%.

The dose mapping of commercial Co60 radiation source installed at PARAS Lahore was carried out by fixing/placing red PMMA sheets (2 mm) as shown in Figure 1 which involved 3 horizontal planes (A, B, and C), each having 15 strips at different locations at the top, middle and bottom of the product box. The dose distribution of the box (container) was carried out with and without the product (dried apricot). The dimensions of the product container used for irradiation were 60x35x40 cm (L × W × H). The box was moved along with other boxes containing medical products into the plant running on normal mode. After irradiation, the absorbance of the red PMMA strips was measured at 640 nm.

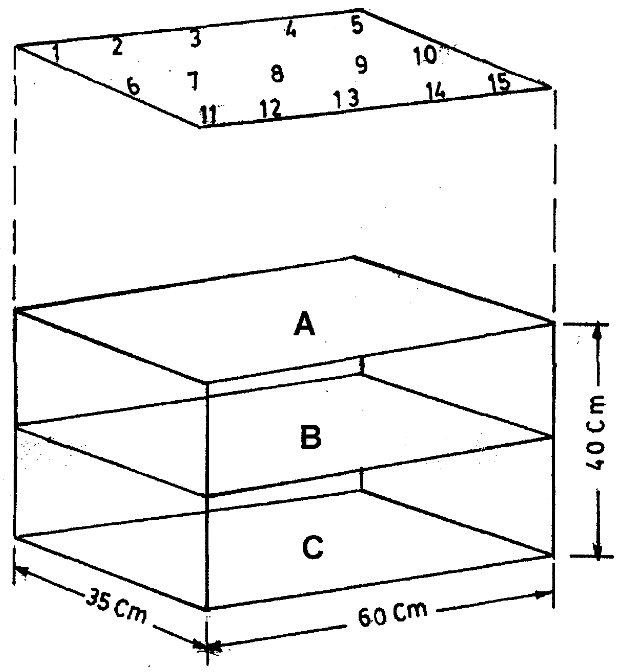

FIG. 1. View of the PMMA sheets in horizontal planes (A, B, C) placed in the box used in PARAS, Lahore.

2.4. Detection tests

2.4.1. Label dosimeters

Radiation dosimetry is fundamental to all field of sciences dealing with ionizing radiation and its effects. Radiation dosimetry is therefore, a key parameter to the technological developments regarding radiation food processing. In order to develop some suitable screening method for detection and measuring the absorbed dose, the following materials were employed:

2.4.1.1. Flexible polymers

Locally manufactured polyethylene in different colors and thickness and clear PVC were irradiated and their absorption was measured at maximum absorption wavelength using UVVis recording spectrophotometer.

2.4.1.2. Rigid polymers

Locally manufactured PMMA in different colors and thickness were tested. The 210 mm thick strips of transparent PMMA were irradiated either with gamma radiation doses ranging from 0.1 to 3.0 kGy or 0.125–1.0 kGy. Initially their spectral absorption over a range of 200 to 1100 nm was measured in order to obtain the maximum peak absorption in each case. Their absorption and/or specific absorption (K) was measured at their maximum peak absorption spectra. The specific absorption is optical absorbance at the measuring wavelength (A) divided by the dosimeter thickness (t). Among the dosimeters, those found suitable (clear PMMA) were tested for their storage stability at ambient room conditions (20–35°C; RH 40–80%).

2.4.1.3. Sterin indicator

Two versions of Sterin indicators were investigated i.e. Sterin 125 and Sterin 300 to provide visual positive verification of irradiation treatment at or above 125 and 300 Gy respectively. This is a new device commercially made available by the International Specialty Products (ISP) Inc. Wayne N. J. USA [18]. It consists essentially of multiple layers of colored and clear films changing color and transparency, respectively, with dose thus creating the dosimetric effect. Sterin indicators are used as quality assurance devices for irradiation disinfestation regarding quarantine regulations. These labels are designed as threshold indicators, where a visual message changes from *NOT* IRRADIATED before exposure to IRRADIATED after exposure at, or above the threshold irradiation dose.

For evaluation purposes, these two types of indicators (Sterin 125 and Sterin 300) were irradiated at dose ranges of 75–175 Gy and 200–400 Gy respectively. The stability of these Sterin indicators was also subjectively evaluated during the following storage conditions:

1. Temperature: ambient room (15–35°C; RH 35–70%)

2. Light:
    - sunlight for 60 hours during December 1997 (15–20°C)
    - fluorescent light (at 100 ft. c; 20–25°C)

The subjective visual evaluation involving 15 judges was made using Multiple Comparison Difference Analysis. A method for Difference Analysis reported by Larmond [19] for foods, was modified by the authors for nonfood materials in scoring difference between irradiated Sterin labels at different doses and untreated product containing the visible impression NOT IRRADIATED. The details of the subjective test involved are shown below:

MULTIPLE COMPARISON DIFFERENCE ANALYSIS

Name: **Date:________________**

Signature:

QUESTIONNAIRE:

You are receiving samples of STERIN INDICATORS to compare for irradiation treatment. You have been given a reference (unirradiated) sample, with the mark: NOT IRRADIATED to which you are to compare each irradiated sample and score the difference using scale 1–9. With irradiation treatment the visibility of the word NOT is changed. Mark the degree of difference that exists with regard to the visibility of word "NOT".

SAMPLE NUMBER

AMOUNT OF DIFFERENCE:

None (1)

Slight (23)

Moderate (45)

Much (67)

Extreme (89)

COMMENTS:

Revised from Larmond, E. [19]. Methods for sensory evaluation of food. Canada Department Agric. Publ. 1284 pp 1924.

2.4.1.4. Collaborative studies

Intercomparison studies were conducted among Pakistan, China and Bangladesh to evaluate the suitability of clear and yellow PMMA as label dosimeters for measuring absorbed doses in fresh/dried fruits and tree nuts. For this purpose, clear PMMA in the thickness of 2, 4, 6, 8 and 10 mm was sent to the counterparts in China and Bangladesh and in return yellow PMMA was received from China which was evaluated by measuring its specific absorption at 530 nm.

2.4.2. Analytical techniques

For thermoluminescence (TL) measurements, the minerals separated from the irradiated and unirradiated samples (15 mg) were placed on to a clean stainless steel disc (10 mm dia. 0.5 mm thickness) and their TL measured according to Khan and Delincee [2] at the temperature range of 80–320^{O}C using TL dosimeter model Harshaw 2000C coupled with automatic integrator 2000B. The chemiluminescence (CL) intensity of as such, moisture free, fat free, charred and ashed samples of fresh fruits, dried fruit and tree nuts were recorded using luminol (5amino2, 3 dihydro1, 4phthalazine dione) and lucigenin (bisNmethyl acridinium nitrate) reactions with the help of luminometer1250 Bio. Orbit (3). The radiolytic changes in fatty acids of irradiated and unirradiated tree nuts were determined by gas chromatography. The oil from tree nuts was extracted using petroleum ether (b.p. 4060°C), esterified (20) and analysed for fatty acids profile using Perkin Elmer model 3920 Gas Liquid Chromatograph.

2.5. Statistical analysis

Wherever applicable, the data were statistically analysed using means, standard deviation (SD), regression/correlation and coefficient of variation (CV) according to Little and Hills [21].

3. RESULTS AND DISCUSSION

3.1 Density measurements

Considerable variation in density was observed depending upon the nature of fruit/nut and the package employed. For overseas trade invariably hardboard containers of 2 sizes are used in Pakistan. However, for incountry marketing, wooden crates and nylon bags are employed. The density of products in hardboard cartons was generally comparable to that in nylon bags while both were lesser than wooden crates because of difference in the weight of the packages and packaging arrangement.

TABLE 1. DENSITY OF FRESH/DRIED FRUITS AND TREE NUTS IN DIFFERENT CONTAINERS

Material	Density (g/cm^3)		
	Hardboard carton	Wooden box	Nylon bag
Fresh fruits			
apple	0.45–0.49	0.52–0.54	0.42–0.44
mango	0.44–0.48	0.56–0.58	0.46–0.48
Dried fruits			
apricot	0.50–0.52	0.64–0.66	0.56–0.59
date	0.55–0.57	0.59–0.60	0.50–0.52
raisin	0.53–0.56	0.66–0.68	0.57–0.59
Tree nuts			
almond	0.46–0.49	0.56–0.58	0.48–0.50
pinenut	0.45–0.47	0.60–0.62	0.49–0.51
walnut	0.40–0.43	0.43–0.45	0.40–0.43

3.2. Depth dose measurements

In the case of NIFA research irradiator, isodose points determined in respect of both the vertical and horizontal planes indicated that the absorbed dose was uniform at the central positions. Minimum dose points were located at the corners of planes. The relative dose in loaded (with apricot) container also revealed the same trend in the absorbed dose distribution within the chamber. However, there was a decrease in the absorbed dose because of the product density. It was found that the depthdose profiles for horizontal planes were similar to that of vertical planes. Parts of the PMMA sheet facing Co60 pencils showed higher doses due to the dose build up effect and emission of secondary electrons as compared with the central zone of the radiation chamber, where the dose distribution was uniform. These isodose curves show the relative position and location of maximum and minimum dose. The data revealed minimum dose (D min) at the top & bottom positions, and maximum dose (Dmax) at the sides of the radiation chamber which were adjacent to the source. There was symmetrical distribution of absorbed dose within the volume of the chamber and it was observed that the central region of the radiation chamber (about 14 × 4 × 6 cm) is quite uniform in dose distribution.

The results on the depth dose measurement of PARAS are given in Table 2. The dose distribution for planes A, B and C showed almost uniform distribution from the top to bottom except minor variations for few strips either at the top, middle or bottom positions. The strips 610 especially 79 at planes A, B and C showed comparatively lesser dose than others places because of their distance from source (see Figure 1). The pattern of dose distribution in the container with the product was similar to that of without product but the values were variably lower. The measurements of means, standard deviations and the coefficient of variability (CV) in relation to the position of the strips and the planes indicated generally a good uniformity in dose distribution within the box.

TABLE 2. DOSE DISTRIBUTION OF CO60 COMMERCIAL IRRADIATOR PARAS, LAHORE)

Positions	WITHOUT PRODUCT						WITH PRODUCT					
	A	B	C	Mean	SD	CV	A	B	C	Mean	SD	CV
1	28.1	28.2	27.7	28.0	0.26	0.94	26.7	26.8	26.3	26.6	0.26	0.99
2	27.9	28.0	27.3	27.7	0.38	1.37	26.2	26.3	25.7	26.1	0.32	1.23
3	27.7	27.8	26.8	27.4	0.55	2.00	258	25.9	24.4	25.4	0.84	3.31
4	27.8	28.1	27.6	27.8	0.25	0.90	26.1	26.4	25.9	26.1	0.25	0.96
5	28.0	28.5	28.2	28.2	0.25	0.89	26.6	27.1	26.8	26.8	0.25	0.94
6	26.0	26.2	25.9	26.0	0.15	0.58	24.7	24.9	24.6	24.7	0.15	0.62
7	25.9	26.2	25.8	26.0	0.21	0.80	23.8	24.1	23.7	23.9	0.21	0.87
8	25.8	26.3	25.6	25.9	0.36	1.39	23.5	23.9	23.3	23.6	0.31	1.29
9	25.9	26.4	25.7	26.0	0.36	1.39	23.8	24.3	23.6	23.9	0.36	1.51
10	25.9	26.5	25.8	26.1	0.38	1.45	24.6	25.2	23.0	24.3	0.11	4.69
11	27.5	28.8	28.1	28.1	0.65	2.31	26.1	27.4	26.7	26.7	0.65	2.43
12	27.5	28.2	28.0	27.9	0.36	1.29	26.1	26.8	26.6	26.1	0.59	2.24
13	27.6	28.5	27.9	28.0	0.46	1.64	25.9	26.8	25.7	26.1	0.59	2.24
14	27.8	28.4	28.0	28.1	0.31	1.09	26.4	27.0	26.6	26.7	0.31	1.15
15	27.9	28.3	28.0	28.1	0.21	0.74	26.5	26.9	26.6	26.7	0.21	0.78
Mean	27.1	27.4	27.0				25.5	25.9	25.3			
SD	0.93	0.95	1.0.3				1.11	1.19	1.38			
CV	3.43	3.45	3.82				4.38	4.58	5.48			

Values are in kGy and A, B, C are the horizontal planes.
Container size 60 × 35 × 40 cm^3. Product: dried apricot.

3.3. Label dosimeters

Suitability of flexible polymers (PE,PVC) was studied to develop a suitable label dosimeter to detect the irradiation treatment. For this purpose the polymers were irradiated in the dose range of 0.1–3.0 kGy and their absorption recorded at the peak absorption wavelengths (Table 3). It was noted that the response was irregular even immediately after irradiation. Therefore, it was considered worthwhile to test the rigid polymers (PMMA) in different colours and thickness, which revealed satisfactory response to irradiation treatment in certain cases especially the red and clear PMMA. The PMMAs in other colours did not give reliable/consistent response.

TABLE 3. ABSORPTION OF IRRADIATED FLEXIBLE PACKAGES

Sample	λ	Radiation dose (kGy)									
	(nm)	0	0.1	0.25	0.5	1.0	1.5	2.0	2.5	3.0	Mean
P.E. clear (L.D)	220	0.36	0.36	0.29	0.30	0.33	0.31	0.41	0.35	0.30	0.33
P.E. clear (H.D)	220	1.96	1.77	1.90	1.90	1.92	1.86	1.81	1.83	1.77	1.85
P.E clear (O.P.P)	220	0.24	0.34	0.35	0.27	0.22	0.21	0.20	0.23	0.23	0.25
PE Yellow	530	2.31	2.29	2.27	2.31	2.27	2.58	2.61	2.22	2.33	2.36
PVCClear	245	1.05	1.11	1.07	0.99	1.01	1.04	0.99	1.00	1.03	1.03
Mean	1.18	1.1	1.18	1.16	1.15	1.20	1.21	1.12	1.13	1.13	

Values are average of 23 determinations.
PE Polyethylene (L.D= low density; H.D.= high density).
PVC Polyvinyl chloride.

Specific absorption (K values) of clear and red PMMA (2 mm) was studied over 2 different storage intervals. In the first case, lower storage duration (42–60 hours) was tried while in the second longer storage periods (1–12 months) were tested and the results of these experiments are shown in Tables 4 and 5 respectively. The specific absorption was almost linear immediately after irradiation. The linearity during successive storage intervals up to 260 hours was found to be stable in both the PMMA tested but the K values of clear irradiated samples decreased consistently with increasing storage. However, the K values of red PMMA (irradiated) remained generally stable for the entire storage of 260 hours. In the second experiment involving the longer storage it was observed that the linearity over a dose of 0.1–3.0 was detectable up to 1 month, beyond which the K values decreased up to 6 months and then slightly increased on 12 months storage probably due to changes in the environmental temperature and humidity in both the clear and in some cases red PMMA. Specific absorption in relation to absorbed dose (0.1–3.0 kGy) of clear and red PMMA (2 mm) did not remain linear on or beyond one month storage. The data indicates that clear and red PMMA (2 mm) can only be used to detect irradiated food up to maximum of 1 month storage at ambient conditions and that the verification of dose >0.5 kGy over a tested range of 0.1–3.0 kGy seems possible.

In view of the limitation in stability of 2 mm PMMA, it was considered important that work on clear PMMA involving thickness up to 10 mm should be conducted in order to achieve longer stability of radiation dosimeter. Therefore, further studies were conducted on the response of clear PMMA in the thickness of 2, 4, 6, 8 and 10 mm and their stability was tested up to 6 months at ambient conditions and the data are presented in Table 6.

The data indicated that optical response of irradiated clear PMMA for all the tested thickness, was almost linear in relation to the absorbed doses immediately after treatment. Although the response decreased slightly during 6 months storage for the dosimeters in the thickness range of 4–10 mm, the values for 2mm PMMA decreased much faster than others.

TABLE 4. SPECIFIC ABSORPTION OF IRRADIATED CLEAR AND RED PMMA (2 mm) DURING SHORT STORAGE

Storage (hour)	Clear PMMA (λ = 320 nm) Irradiarion Doses (kGy)					Red PMMA (λ = 600 nm) Irradiation doses (kGy)				
	0	0.1	3.0	Mean	CV	0	0.1	3.0	Mean	CV
0	0.045	0.050	0.099	0.06	46.1	0.145	0.141	0.272	0.168	40.1
4	0.045	0.051	0.092	0.06	44.2	0.145	0.143	0.296	0.195	44.1
20	0.043	0.051	0.097	0.06	45.8	0.128	0.144	0.307	0.193	51.3
60	0.039	0.044	0.083	0.05	43.5	0.142	0.138	0.306	0.195	49.1
100	0.040	0.043	0.081	0.05	41.8	0.139	0.137	0.299	0.192	48.5
120	0.033	0.038	0.073	0.048	45.4	0.136	0.136	0.284	0.185	46.1
140	0.038	0.044	0.079	0.05	41.3	0.139	0.137	0.289	0.188	46.3
260	0.037	0.042	0.074	0.051	39.4	0.140	0.139	0.250	0.176	36.2
Mean	0.04	0.050	0.09			0.139	0.139	0.288		
CV	16.4	10.5	12.6			3.9	2.1	6.7		

Values are averages of 23 determinations.

TABLE 5. SPECIFIC ABSORPTION OF CLEAR AND RED IRRADIATED PMMA DURING LONG STORAGE

Storage (months)	Radiation dose (kGy)							
	0	0.1	0.5	1.0	1.5	2.0	3.0	Mean
Clear PMMA (320 nm)								
0	0.03	0.04	0.05	0.06	0.07	0.08	0.09	0.06
1	0.03	0.04	0.04	0.05	0.06	0.07	0.08	0.06
6	0.04	0.04	0.05	0.05	0.05	0.05	0.05	0.05
12	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05
Mean	0.04	0.04	0.05	0.06	0.06	0.06	0.07	
Red PMMA (600 nm)								
0	0.19	0.18	0.25	0.29	0.37	0.46	0.55	0.33
1	0.14	0.14	0.21	0.22	0.23	0.24	0.25	0.20
6	0.15	0.15	0.16	0.16	0.16	0.17	0.17	0.16
12	0.19	0.18	0.19	0.18	0.18	0.18	0.18	0.18
Mean	0.17	0.17	0.20	0.21	0.24	0.24	0.26	

Values are average of 23 determinations.

TABLE 6. EFFECT OF IRRADIATION ON ABSORBANCE AND STABILITY OF CLEAR PMMA

	Storage months						
DOSE kGy	0	1	2	3	4	5	6
				2 mm			
0.125	0.003	0.003	0.003	0.050	0.014	0.090	0.070
0.25	0.011	0.010	0.010	0.010	0.016	0.098	0.077
0.50	0.014	0.014	0.011	0.010	0.010	0.093	0.075
0.75	0.023	0.020	0.019	0.012	0.013	0.099	0.098
1.00	0.035	0.021	0.020	0.012	0.011	0.103	0.122
				4 mm			
0.125	0.012	0.012	0.011	0.015	0.015	0.020	0.021
0.25	0.029	0.014	0.012	0.019	0.015	0.022	0.022
0.50	0.051	0.028	0.015	0.019	0.017	0.021	0.023
0.75	0.068	0.034	0.029	0.027	0.022	0.026	0.022
1.00	0.091	0.053	0.031	0.031	0.027	0.034	0.036
				6 mm			
0.125	0.015	0.012	0.010	0.012	0.0123	0.025	0.024
0.25	0.018	0.015	0.020	0.018	0.018	0.020	0.022
0.50	0.062	0.053	0.047	0.031	0.031	0.024	0.024
0.75	0.105	0.089	0.080	0.064	0.064	0.048	0.044
1.00	0.126	0.108	0.093	0.069	0.069	0.055	0.057
				8 mm			
0.125	0.030	0.026	0.040	0.035	0.035	0.037	0.037
0.25	0.053	0.051	0.055	0.049	0.048	0.053	0.054
0.50	0.125	0.100	0.088	0.075	0.075	0.053	0.055
0.75	0.162	0.121	0.114	0.099	0.099	0.085	0.094
1.00	0.202	0.149	0.136	0.128	0.128	0.113	0.111
				10 mm			
0.125	0.039	0.038	0.042	0.040	0.049	0.040	0.041
0.25	0.061	0.074	0.084	0.085	0.090	0.085	0.079
0.50	0.146	0.133	0.126	0.124	0.131	0.124	0.112
0.75	0.206	0.191	0.181	0.177	0.165	0.160	0.160
1.00	0.242	0.236	0.220	0.210	0.200	0.198	0.194

Values are average of 3 determinations taken at 320 nm.

The present studies showed that clear PMMA from 4–10 mm (especially 8–10 mm) in thickness can serve as a useful and reliable routine dosimeter for irradiated food materials in the dose range 0.125–1.00 kGy.

3.4. Intercomparison studies on pmma

In order to evaluate the suitability of clear PMMA as a label dosimeter, 2–10 mm thick PMMA sheets were sent to China and Bangladesh for interlaboratory study. In return, yellow PMMA strips were received from China which were evaluated in the dose range of 125 to 1000 Gy of gamma radiation at NIFA and the results are shown in Table 7. The data revealed a linear increase in specific absorption of the yellow PMMA with the increase in irradiation dose. It was therefore, concluded that the YLPMMA developed by the Institute for Application of Atomic Energy, Beijing, China can be successfully used in detecting irradiation treatment of dried fruits and tree nuts in the range of radiation doses studied (125–1000 Gy).

TABLE 7. EFFECT OF IRRADIATION DOSE ON SPECIFIC ABSORPTION OF YL PMMA

Irradiation dose (Gy)	Specific absorption measurements		Mean ± SD
	1	2	
125	0.1030	0.1020	0.1025 ± 0.0005
250	0.1680	0.1710	0.1695 ± 0.0015
500	0.3250	0.3490	0.3370 ± 0.0020
750	0.5710	0.6510	0.6110 ± 0.0400
1000	0.8640	0.8760	0.8700 ± 0.0060

TABLE 8. SUBJECTIVE EVALUATION OF IRRADIATED *STERIN* INDICATOR

Samples	Mean score of 15 judges					
	Storage months					Exposure hours
Dose Gy		Ambient dark		Fluorescent light		Sunlight
	initial	6	12	6	12	60
Sterin125						
NOT Irradiated	1.0	1.0	2.0	2.0	3.0	3.0
75	4.3	5.0	5.9	6.3	7.1	7.6
100	6.1	7.2	7.9	7.5	8.1	7.8
125	8.0	8.0	8.5	8.4	8.7	8.5
150	8.4	8.5	8.7	8.6	8.7	9.0
175	8.5	8.6	8.6	8.6	8.7	9.0
Sterin300						
NOT Irradiated	1.0	1.0	2.0	2.0	3.0	3.0
200	4.5	6.1	6.3	6.6	7.2	7.9
250	6.8	7.2	7.3	7.3	8.6	8.6
300	8.4	8.6	8.7	8.9	9.0	9.0
350	8.8	8.8	8.6	8.8	9.0	9.0
400	8.2	8.5	8.6	8.9	9.0	9.0

SD range = ±0.23–0.55 for 15 judgements.

As for the clear PMMA (210 mm) sent from Pakistan to Bangladesh, preliminary information received indicated that the response of the 4–8 mm is fairly good in the 200–2000 Gy of gamma radiation.

3.5. Evaluation of sterin indicator

The sets of 6 labels for each sterin indicator (Sterin 125 and Sterin 300) irradiated at different doses were presented to 15 judges in individual sessions with the task to rank them according to the extent of difference as compared to the untreated Sterin labels showing the mark *NOT*. The subjective analyses were made for both types of Sterin indicators kept at different storage conditions. The results are presented in Table 8 which showed obvious changes and the judges were able to discriminate between untreated and treated samples. The subjective evaluation indicated that these indicators are fairly reliable at and above the designated doses, however, they were also affected by the dose lower than their threshold values. It shows that there is a need of developing indicators which show abrupt change in optical properties at the declared threshold dose. Lower scores were given to the sterin samples irradiated at suboptimal doses because the word *NOT* was not completely invisible. It was observed that during advanced storage under ambient dark, fluorescent light and sunlight, the darkshades of irradiated samples further darkened. However, it was interesting to find that even the untreated samples showing the mark *NOT* also slightly darkened under fluorescent and sunlight conditions and the dark shade increased as the storage advanced, but still the word *NOT* remained visible. Similar observations have been reported by other workers [6, 7]. The slight darkening of the red field upon extended exposure to fluorescent and sunlight warrants that these indicators must be sealed inside the opaque or impermeable sachets.

3.6. Analytical tests

The effect of radiation doses (0.51–.5 kGy) on the TL values of minerals obtained from the dried fruits and plant nuts was studied at different temperatures. The maximum peak values of the glowcurves were observed at 200°C which are presented in Table 9.

TABLE 9. THERMOLUMINESCENCE VALUES (nC) OF INORGANIC PARTICULATES FROM IRRADIATED DRIED FRUITS/NUTS AT 200°C

Dose (kGy)	Samples					
	Apricot	Date	Raisin	Almond	Pinenut	Walnut
0	3.2	17.7	25.3	25.3	3.2	8.3
0.5	1025.2	3457.4	2163.9	2952.7	1187.1	1560.5
1.0	1051.5	5849.7	3020.7	4882.2	2281.1	3308.2
1.5	2013.9	8540.9	4203.2	9693.5	7151.1	3824.4
Correlation Coefficient (r)	0.98	0.99	0.98	0.98	0.97	0.99

The TL values at each temperature increased with increasing radiation absorbed dose in the products. There was a manifold increase in the values on irradiation and the increase was linear in relation to the dose applied. However, the TL values obtained from several fruits/nuts were different, but the change in the glow pattern was identical between samples. The test statistics indicated the presence of significant linear regression ($P < 0.01$) and the good to strong correlation between TL values and radiation doses were observed for all the dried fruits and plant nuts. The difference in TL efficiency may be attributed to differences in dust composition as in all cases equal amount of dust particulate minerals (15 mg) were used for TL measurements. It was therefore concluded that as the absorbed radiation doses were increased, the TL values increased at each temperature, and at the peak TL values (200 °C) for each dried fruit/nuts, the relationship was expressed by significant linear regression of the forms:

$Y= 158 + 1296.9$ x (apricot)
$Y= 269 + 5593.4$ x (date)
$Y= 319 + 2690.6$ x (raisin)
$Y= 252 + 6144.6$ x (almond)
$Y= 388 + 5468.2$ x (pinenut)
$Y= 135 + 2453.0$ x (walnut).

Earlier reports claimed that TL is one of the several techniques recommended for adaptation as reliable and simple identification method for herbs, spices and fruits [2, 22, 23]. Some other workers [24], reported that initial fading of TL signals was rapid for all the spices but the TL response of irradiated spices persisted above those of unirradiated ones for over 2 months after irradiation. GoeksuOgelman and Regulla [25] indicated that inorganic dust on spices was responsible for TL emissions. In another study, Bogl [22] observed that TL was a rapid method for identification of irradiated dried food during long term experiments. He also suggested that for many spices, an identification by this technique was possible as late as one year after irradiation or even later. Heide et al. [24] while detecting irradiated strawberries by TL measurements found that TL values of irradiated and unirradiated samples increased with the increase in the voltage of the photomultiplier tube, however, the optimizing ratio (TL of irradiated/TL of unirradiated sample) was not affected by voltage. They further noticed that as the intensities were dependent on the amount of adhering minerals and the radiation doses, the dose response curves of whole sample measurements could not be completely reproduced.

The CL emissions of dried fruits/nuts having notably high moisture and fat contents, was measured on as such, moisture free and fat free basis, as a result of luminol and lucigenin reactions. The comparison of peak height (cm) values showed wide variation among the samples. It was observed that radiation treatment did not influence the CL intensity to any level of practical interest. Strangely large consistent increases in the CL response were observed in the ashedmaterial of irradiated samples than the unirradiated controls and the reason for these increases is not clear. The data on fatty acids indicated that most of the fatty acids were not affected by a dose of 1.5 kGy gamma radiation and there was an irregular influence of radiation treatment. There was a prominent decrease in the lauric acid content of apricot's stone while increases in the lauric acid and linolenic acid in pinenut oil upon irradiation were noted.

4. CONCLUSIONS

It was concluded that density varied depending upon the package size, type and the nature of the fresh or dried fruit/nut. Among the label dosimeters the samples of clear PMMA, in the thickness range of 4–10 mm (especially 8–10 mm) gave a linear response in relation to the irradiation doses and the optical response was stable almost up to 6 months tested storage. As for the Sterin indicators, their subjective evaluation revealed that this new material is fairly reliable but is affected by doses lower than the designated threshold and exposure to light. The YLPMMA dosimeter developed by China was useful in the range of 125–1000 Gy of gamma radiation. Dose distribution studies of research irradiator at NIFA and a commercial gamma source (PARAS), at Lahore indicated generally a good dose uniformity in their product containers. Among the analytical tests, TL measurements gave clear and reproducible differences between irradiated and unirradiated samples and this technique can be used to measure absorbed dose in dried fruits/tree nuts.

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REFERENCES

[1] IAEA, Analytical detection methods for irradiated food. IAEA-TECDOC-587 (1991) p.8.
[2] Khan, H.M., Delincee, H., Detection of irradiation treatment of dates using thermoluminescence of mineral contaminants. Rad. Phys. Chem. **46** (1995): 717–720.
[3] Sattar, A., Delincee, H., Diehl, J. F., Detection of gamma irradiated pepper and papain by chemiluminescence. Radiat. Phys. Chem. **29** (1987): 215–221
[4] Sattar, A., Ahmad, A., Atta S., Ahmad T., Development of detection methods and label dosimeters for gamma irradiated dried fruits and tree nuts. " In: Impact of food research on new product development". (Ali, R., Barlow, P.J. Whitaker, J. R., Ed.). Proc. 2nd Inter. Conf. Uni. Karachi, Pakistan (1995) 97–104.
[5] Ahmad, A., Sattar, A., Iqbal, M. Z., Detection of irradiated dried fruits and plant nuts by chemiluminescence measurements. Pak. J. Food Sci. **5** (1995) 23–25.
[6] Razem, D., Dosimetric performance of and environmental effects on Sterin irradiation indicator labels. Radiat. Phys. Chem. **49** (1997). 491–495.
[7] Ehlermann, D.A.E., Validation of a label dosimeter for food irradiation applications by subjective and objective means. Appl. Radiat. Isot**. 48** (1997). 1197–1201.
[8] Sattar, A., Ahmad, A. Atta, S., Development of detection methods and label dosimeters for gamma irradiated dried fruits and tree nuts. The impact of food research on new product development (Ali, R., Barlow, P.J. Whitaker, J.R., Eds). Proc. 2nd Intern. Conf., Karachi, Pakistan 28–31 Jan. (1995) 97–104.
[9] Sattar, A., Ahmad, A., Iqbal, M.Z., Identification of irradiated dried fruits and plant nuts by different analytical techniques. Pak. J. Food Sci. **5** (1995) 69–72.
[10] Sattar, A., Ahmad, A., Iqbal, M.Z., Identification of irradiated dried fruits, tree nut in trade. Proc. Nat. Conf. Postharvest Tech. Horticultural commodities 1012 Sep FAO Quetta (1997) 94–100.
[11] Ahmad, A., Sattar, A., Hussain, A. Iqbal, M.Z., Effect of irradiation on lipid characteristics of plant nut oils. Sains Malaysiana. **26** (1997).
[12] Sattar, A., Ahmad, A., Atta, S., Evaluation of different techniques to identify irradiated meats, Chicken and Fish. In impact of food research on new products development (Ali, R., Barlow, P.J. Whitaker, J.R., Ed) Proc. 3rd Intrn. Conf. 36 June, Uni. Karachi (1997).
[13] Sattar, A., Ahmad, A., Iqbal, M.Z., Analytical detection methods for gamma irradiated fresh apples. Pak. J. Food Sci., **7**: 1997 pp 5355
[14] Ahmad, A., Sattar, A., Ali, I., Iqbal, M.Z., Irradiation effect on thermoluminescence values in dried fruits. J. Radio Anal. & Nucl. Chem. **227** (1998) accepted.
[15] Ahmad, A., Sattar, A., Ali, I., Iqbal, M.Z., Effect of gamma irradiation on thermoluminescence emissions of Asian plant nuts. Die NahrungFood **43:** (1999) accepted.
[16] Government Pakistan, Pakistan Atomic Energy Notification. The Gazette of Pakistan, Government of Pakistan. Islamabad March 7 (1996) 339–345.

[17] Fricke, H., Hart, H.J., Chemical Dosimetry. In Radiation dosimetry (Attix, F.H., Roesch, W.C., Eds) Academic Press N.Y. (1966).
[18] Lewis, D.F., Listl, C.A., Radiation dose indicator. US patent No. 5084623 (1992).
[19] Larmond, E., Methods for sensory evaluation of food. Canada Dept. Agric. Pub. 1284 (1977). 19–24.
[20] Shehata, A.Y., deMan, J.M., Alexander, J.C., A simple and rapid method for preparation of methyl esters of fats in milligram amounts for gas chromatography. Can. Inst. Food Technol. J. **3** (1970) 85–90.
[21] Little, J.M., Hills, F.J., Statistical methods in agriculture research; Agric. Extension Univ. California Davis USA (1972) 121–137.
[22] Bogl, K.W., Methods for identification of irradiated food. Rad. Phys. Chem. **35** (1990). 301–310.
[23] Moriarty, T.F., Oduko, J.M., Spyrou, N.M., Thermoluminescence in irradiated food stuffs. Nature **332** (1988) 22.
[24] Heide, L., Guggenberger R., Bogl, K.W., Application of thermoluminescence measurements to detect irradiated strawberries. J. Agric. Food chem. **29** (1990) 2160–2163.
[25] Goksu Ogelman, H.Y., Regulla, D.F., Detection of irradiated food. Nature **340** (1989) 23.

PROCESS CONTROL AND DOSIMETRY APPLIED TO ESTABLISH A RELATION BETWEEN REFERENCE DOSE MEASUREMENTS AND ACTUAL DOSE DISTRIBUTION

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Abstract. The availability of the first commercial dose level indicator prompted attempts to verify radiation absorbed dose to items under quarantine control (eg for insect disinfestation) by some indicator attached to these items. Samples of the new commercial dose level indicators were tested for their metrological properties using gamma and electron irradiation. The devices are suitable for the intended purpose and the subjective judgement whether the threshold dose was surpassed is possible in a reliable manner. The subjective judgements are completely backed by the instrumental results. Consequently, a prototype reader was developed; first tests were successful.

The value of dose level indicators and the implications of its use for food or quarantine inspection depends on a link between dose measured (indicated) at the position of such indicator and the characteristic parameters of the frequency distribution of dose throughout the product load ie a box or a container or a whole batch of multiple units. Therefore, studies into variability and statistical properties of dose distributions obtained under a range of commercial situations were undertaken. Gamma processing at a commercial multipurpose contract irradiator, electron processing and bremsstrahlung applications at a largescale research facility were included; products were apples, potatoes, wheat, maize, pistachio. Studies revealed, that still more detailed information on irradiation geometries are needed in order to render meaningful information from dose label indicators.

1. INTRODUCTION

The final goal of this CRP is to establish a reliable link between a radiation absorbed dose documented outside a container and the dose distribution effected throughout the product load contained therein. The availability of such information is thought to facilitate international trade in radiation processed food. This aspect is extremely important for example in instances where the objective of radiation processing is to fulfill quarantine requirements as for fresh and dried fruits and for tree nuts. For reliable disinfestation, the insects may survive a radiation treatment and may even be able to fly but must not be capable of proliferation. The necessary radiation dose for fruit fly eradication is typically 150 Gy; however, to prevent adult emergence altogether, including the emergence of normal appearing, sterile adult insects capable of flight the generic minimum radiation dose should be 250 Gy for any species of fruit flies. Thus, the achievement of a minimum dose once set by authorities is crucial and its verification is indispensable.

Such information attached to the container could be associated, of course, with the shipping documents; preferably it could be a 'label dosimeter' (not a label indicator) permanently affixed to the container before radiation processing. This dosimeter should directly indicate the dose figure or should also be readable by some hand-held instrument. Both types are likely to be available in the near future; one prototype was included in the programme of this CRP.

The main effort of this contribution was devoted to characterization of dose distributions likely to occur under commercial practices for fruit, dried fruit and nut disinfestation by radiation processing. During the course of this CRP some label dosimeters became available and studies into their properties were added; finally, a prototype reader for such labels was developed. Thus, Part B and C below is only a supplement to the main study; a paper on the validation of some label dosimeters by subjective and objective means was already published elsewhere. Preliminary results had already been reported by participants to the Second Research Coordination Meeting of this CRP in 1996.

2. MATERIALS AND METHODS IN GENERAL

Irradiation was done with a gamma cell (cobalt-60, dose rate 0.5 kGy/h) and a linear accelerator for electrons (indirect mode (microwave) 10 MeV, 10 kW, instantaneous dose rate 10^8 Gy/s, pulse duration 12 ìs; bremsstrahlung (X rays) at 5 MeV, dose rate about 10^4 Gy/s; average dose rates depending on scanning width, repetition rate and conveyor velocity). For experiments on larger scale a commercial gamma facility was available. Treatment was always at ambient temperature and humidity. In order to cover the full dose range and sensitivity of available dosimetry films (GAFchromic DM100 and FarWest FWT6000, sealed in paper/plastic envelopes) the target doses were chosen accordingly (eg minimum dose at 5 kGy); the results can easily be transformed to the dose range of interest (eg 250 Gy) as the geometry and, hence, the dose patterns and the relative dose distribution are not affected by setting of exposure time. Film readings were taken with a colorimeter (transmission mode, CIBA CORNING 257 or FarWest Radiachromic Reader) and filters for the appropriate wave lengths. Reference dosimetry for film calibration was done by use of Fricke dosimeter cross-checked by alanine dosimetry (IDAS) at accelerator and gammacell. In the experiments in cooperation with a contract irradiator Harwell Amber 3042 and spectrophotometer readout were applied in addition to our own systems.

Also, in order to study a larger variety of geometries and bulk densities wheat, maize and potatoes were included as models for fresh and dried fruits as well as tree nuts; one study included apples on commercial cardboard trays stacked on standardized pallets; the main studies were executed on pistachio in commercial cardboard boxes containing 40 or 48 pouches, respectively, 125 g each.

SAS (Statistical Analysis System, Carry, USA) was applied for analysis of data and graphical presentation of results.

3. PART A: GENERAL ASPECTS OF PROCESS CONTROL AND DOSIMETRY

For their purposes, the radiation processing industry has already established and standardized the practice of dose mapping and process validation for each individual item accepted for treatment. This, together with records of process control measures taken, of dose measurements executed during the process, and of observations at critical control points established for the process, renders a bulk of data on random fluctuations of the process. In commercial radiation processing it is to be expected that repeating loading patterns occur regularly which can be characterized for all random fluctuations which are likely to occur and can be related to achievable dose distributions and to the measured dose at some established reference position. A study of such informations can reveal whether verification of absorbed dose administered is possible and reliable in trade. As a model, certain geometries (source product geometries) were studied for electron, bremsstrahlung and gamma ray processing.

3.1. Materials and methods

3.1.1. Gamma ray processing and products irradiated

In cooperation with a contract irradiator, a multipurpose type facility designed for source overlap was used. It could hold two pallets (one in top of the other) of dimensions 1.8 m high, 1.2 m wide and 1.0 m deep (or 2.16 m^3). The carriers moved through 4 positions around the source and were irradiated from 4 sides. Consequently, for some loading patterns the position of the minimum dose values is to be expected in the interior and not at the surface (as for other designs of irradiation facilities); this implies that the expected position of the minimum dose is not accessible for measurements during production runs. The carrier was loaded with one pallet containing 1.7 t of maize and a second pallet containing 0.5 t of apples. As the residence time had to be equal for both loads, no target dose was preset for each treatment; instead, a nominal dose suitable for both dosimeter types utilized in this study was chosen. Apples came on commercial cardboard trays and were stacked into the carrier; 4 trays per layer and 12 layers high (overall density 0.21 g/cm^3). Maize was filled in cardboard boxes (20 × 30 × 40 cm^3), 8 boxes per layer and 8 layers high (overall density 0.80 g/cm^3). Apples were in the upper, maize was in the lower hold of the carrier.

3.1.2. Electron/bremsstrahlung processing and products irradiated

The electron accelerator of the institute (technical details see above) was used in direct and in bremsstrahlung conversion mode. The products were usually put on aluminium trays (40 cm by 40 cm), the filling height depending on product density (ie 4.5 cm height for water equivalent material and 10 MeV electron treatment). Wheat and maize were filled in cardboard boxes fitting on the trays of the conveying system (40 × 40 cm^2 base area, layer thickness of 5.5 cm, corresponding to the range of 10 MeV electrons at a bulk density of about 0.8 g/cm^3). This arrangement was also intended to simulate a possible bulk flow processing, ie a continuously flowing bed (40 cm wide, about 5.5 cm high) in a direction perpendicular to the beam. Potatoes were placed on the trays in a single layer about 4 cm high for one-sided and in a double layer about 7 cm high for double-sided irradiation. In the latter arrangement, vacuum packaging in plastic bags was applied in order to secure maintenance of positions during turning of the stack in double-sided irradiations. For pistachio commercial size boxes (15 × 25 × 40 cm^3) were used containing 40 bags, 125 g each in 4 layers with two rows (the height of 15 cm at a density of 0.35 g/cm^3 is slightly more than the range of 10 MeV electrons); double-sided irradiation was applied for 10 MeV electrons and single-sided irradiation for X rays. In the repetition experiment on pistachio commercial size boxes (17 × 27 × 40 cm^3) of another supplier were used containing 48 bags, 125 g each in 6 layers with two rows (the height of 17 cm at a density of 0.33 g/cm^3 is equivalent to the previous series of experiments with regard to electron penetration).

3.1.3. Dosimetry

All dosimeters were placed strategically in order that each represented equal surface areas, volumes or masses, depending on whether the treatment purpose was surface or volume irradiation. This was achieved by placing dosimeters in the centre of each subvolume they represented for bulk goods or by placing 4 dosimeters in equal distances to each other on the surface of each tuber or fruit. In the case of the commercial gamma-irradiation facility, the operator in addition to the experimental dosimetry used his standard dosimetry system at the established dose mapping positions. Depending on the dosimetry system used, target dose and exposure, respectively, were chosen accordingly in order to use the full range of sensitivity of the particular dosimetry system.

3.1.4. Ddata analysis

Scale transformation and comparison with the Gaussian distribution was used to visualize inherent properties of the respective frequency distributions. 'Standardized normal distribution' is always characterized by a mean of zero and a standard deviation of one; thus, size of the standard deviation in relation to the mean value (ie 'standard error') does not play any role. Cumulative frequency distribution is the integral of the Gaussian bellshaped curve and shows a typical sigmoid pattern. Once the cumulative frequency is converted into probit units this sigmoid curve is converted into a straight line. By this scale transformation any deviation of the measured distribution from the normal distribution becomes more obvious. For ease of understanding the vertical scale in probit units is labelled instead with the corresponding percent cumulative frequency. In the horizontal grid, dotted lines parallel to the 50% or meanline indicate 1, 2 and 3 standard deviation widths around the mean. The horizontal dose scale is easily rescaled from standardized units to the real dose values and equal standard deviations then are represented by equal slopes.

Calculated frequency distributions in these experiments are area, volume and mass density functions. For gamma processing each dosimeter represented 9.7 kg of a total of 1,700 kg of maize and each set of 4 dosimeters represented (the surface of) 2.6 kg of a total of 500 kg of apples. For maize the dosimeters were put in gelatine capsules and centred for the volumes they represented during filling of the boxes. For apples the 4 dosimeters of a set were attached to the surface of the fruit in equal distance to each other, 4 apples prepared this way at representative positions on each tray; the individual orientation of the spiked fruit was at random, the main portion of the apples remaining undoped with dosimeters. For electron processing each dosimeter represented 49 g of wheat and 51 g of maize, respectively. The dosimeters were placed in 5 layers in 1 cm distance, 25 dosimeters in a five by five arrangement per layer. For potatoes 4 dosimeters were attached at equal distances on the surface of each tuber, arrangement and orientation of the individual tubers on the tray was at random. For pistachio (except the dosemapping study) dosimeters were placed at the centre of one side of each bag and on the upper and lower inner surface of the cardboard box. All electron treatments were from top, for double-sided treatment the boxes were turned around an axis parallel to the direction of the scan.

3.2 Results and discussion

3.2.1. Gamma processing of maize and apples

Both dose distributions for maize (Fig. 1) and apples (Fig. 2) irradiated at the commercial facility show great similarity and do not deviate too much from the normal distribution. The strategy in placing dosimeters was different for the experiments reported here (FRCN in tables) and for the mapping effort by the contract irradiator. This is reflected most prominently by the differences in the reported mean dose and extreme values for both experiments. The rather high max/min dose ratio for maize (Table 1) is caused by the intentional overloading of the carriers; partial loading as usual practice would have resulted in a value below 2. The difference of results for maize compared to apples is easily explained by the difference in bulk density (0.8 to 0.2 g/cm^3) and equal exposure to the radiation field.

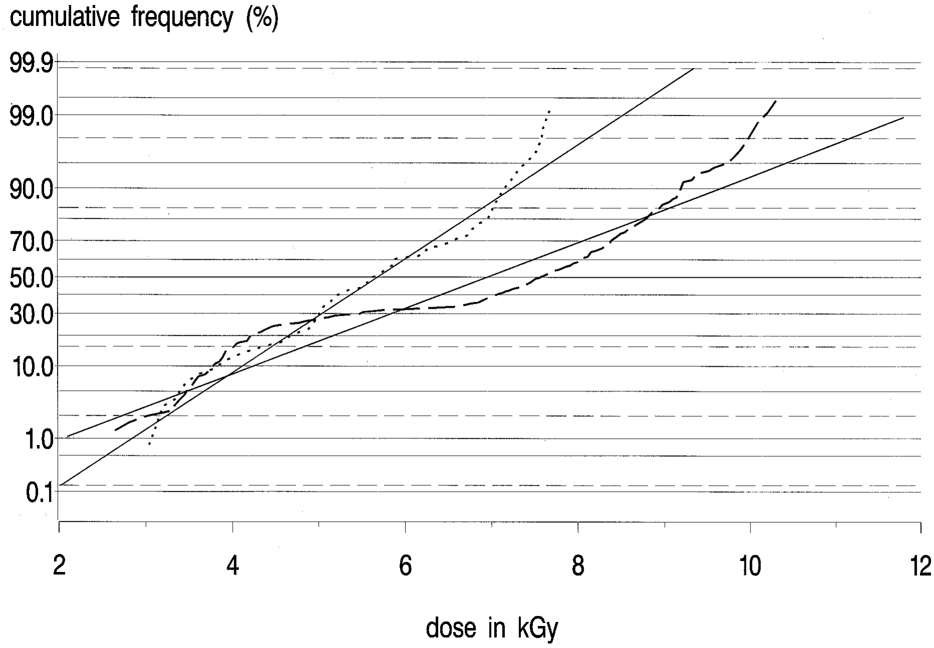

FIG. 1. Gamma processing of maize: frequency distribution of dose determined by FRCN (dotted line) and contract irradiator (dashed line); fine vertical lines: respective mean values.

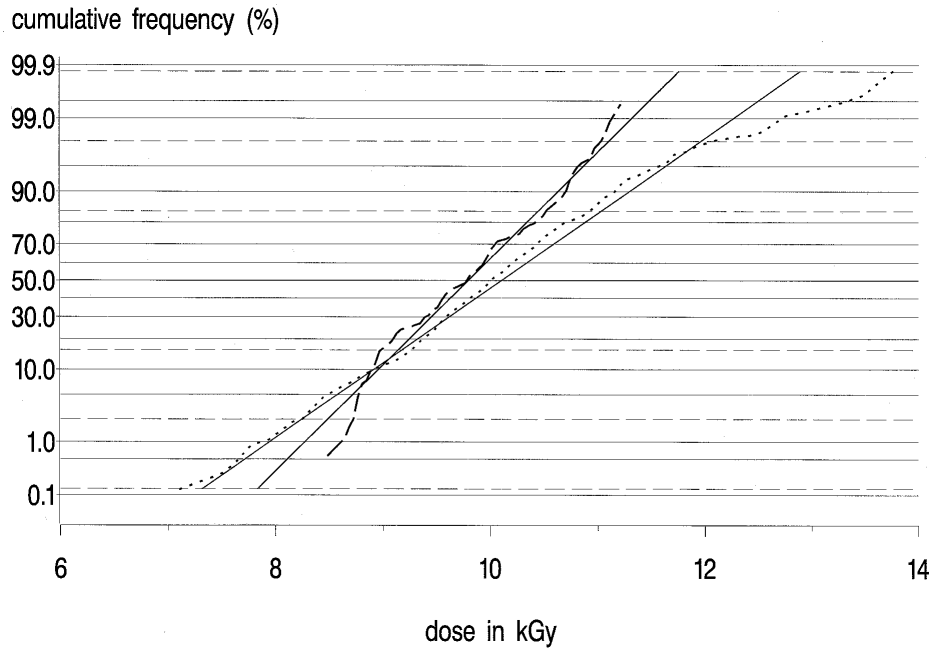

FIG. 2. Gamma processing of apples: frequency distribution of dose determined by FRCN (dotted line) and contract irradiator (dashed line); fine vertical lines: respective mean values.

TABLE 1. CHARACTERISTICS OF DOSE DISTRIBUTION IN GAMMA PROCESSING (dose in kGy)

	mean	std.	min	max	max/min
maize by FRCN	5.68	1.22	3.06	7.84	2.56
by contract irradiator	6.95	2.10	2.70	10.4	3.85
apple by FRCN	9.79	0.652	8.50	11.2	1.32
by contract irradiator	10.1	0.927	7.10	14.2	1.97

3.2.2. Electron processing of wheat, maize and potatoes

Dose distributions for wheat and for maize (Fig. 3) are very similar (Table 2); this was to be expected as geometry (size of boxes and density) were practically identical. The extremely high ratio of max/min dose is caused by the limited range of electrons. For improved homogeneity only layer thickness of about 37 mm instead of the 55 in these experiments would be acceptable. For potatoes (Fig. 4 and Table 3) the relative width of the dose distribution (standard deviation divided by mean dose) is significantly reduced by double-sided irradiation. In order to match the sensitivity of the dosimeter films used the target (surface) dose for all accelerator settings was chosen at 1 kGy, this resulted in higher minimum doses as needed for sprout inhibition. For this reason and for comparison all data were normalized to an equal minimum dose of 0.1 kGy also. The max/min dose ratios (Table 3) show that with 10 MeV electrons processing of a product like potatoes is not possible within acceptable dose ranges; even two-sided irradiation resulted only in insufficient improvement. For example, treating fruits for insect eradication with the recommended dose of 0.3 kGy would result in a maximum dose of about 4.5 kGy which would cause severe radiation damage for various fruits.

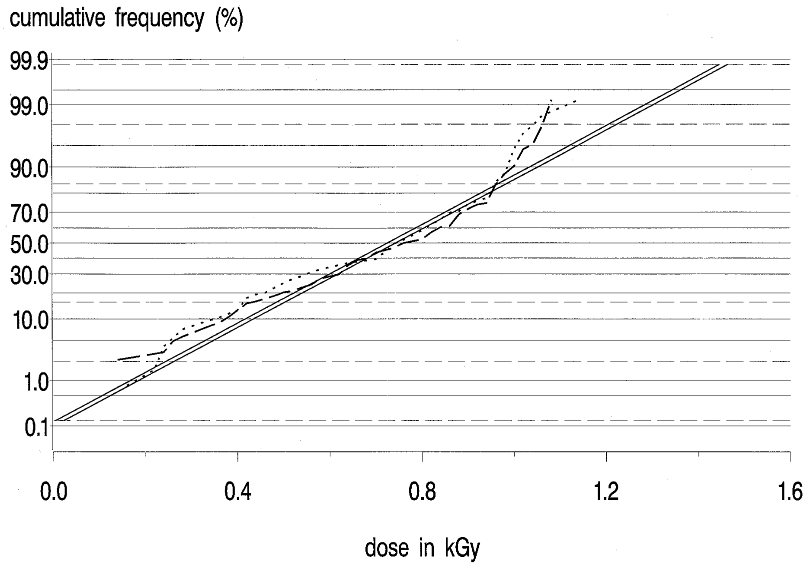

FIG. 3. Electron processing of wheat (dashed line) and maize (dotted line): 10 MeV electrons, single-sided irradiation; fine vertical lines: respective mean values.

TABLE 2. CHARACTERISTICS OF DOSE DISTRIBUTION IN SINGLE-SIDED ELECTRON PROCESSING OF MAIZE AND WHEAT (dose in kGy)

	mean	std.	min	max	max/min
maize	0.743	0.240	0.141	1.15	8.16
wheat	0.725	0.240	0.178	1.19	6.68

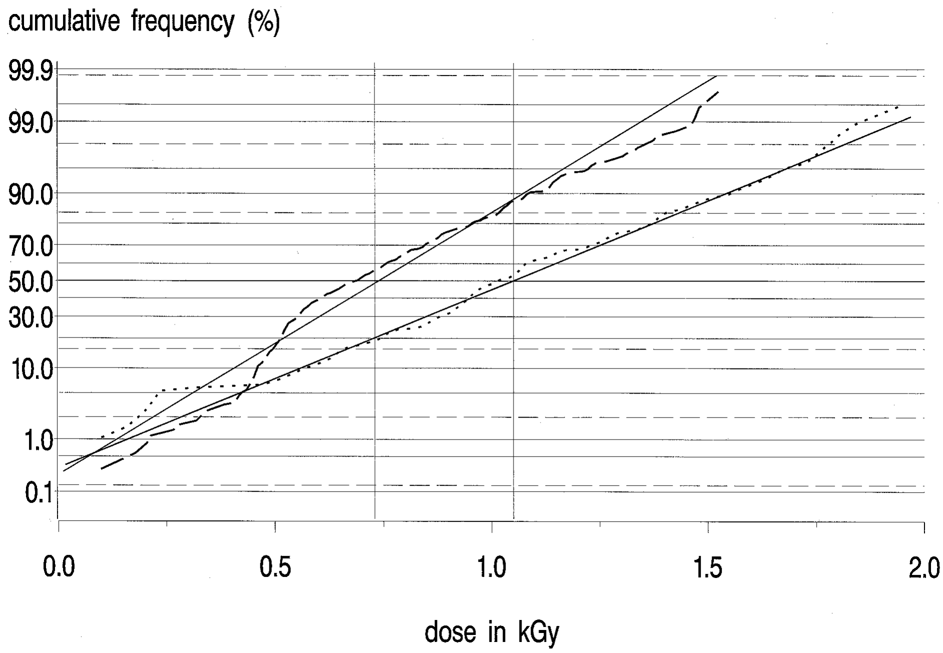

FIG. 4. Electron processing of potatoes: 10 MeV electrons, single-sided (dotted line) and double-sided (dashed line) irradiation; fine vertical lines: respective mean values.

TABLE 3. CHARACTERISTICS OF DOSE DISTRIBUTION IN SINGLE AND DOUBLE-SIDED ELECTRON PROCESSING OF POTATOES (dose in kGy)

	mean	std.	min	max	max/min
single sided	1.07	0.390	0.102	2.07	20.3
double sided	1.67	0.587	0.226	3.48	15.4

3.2.3. Eelectron and bremsstrahlung processing of pistachio

The characteristics of dose distribution for all three variants (including the repetition of the experiment with a larger number of passes and dosimeters) were similar (Table 4); single-sided electron treatment was not applied as commercial cardboard containers were too thick in beam direction for electron penetration (height 15 cm at density of 0.35 g/cm^3 results in an water equivalent thickness of 5.25 cm; practical range of 10 MeV electrons in water is 45 mm). Double-sided electron treatment is in general equivalent to one-sided bremsstrahlung treatment at either 5 or 10 MeV nominal energy. There is no significant difference between the two bremsstrahlung treatments. No extreme overdosing was observed at the surface due to any unfiltered low energy portion of the bremsstrahlung spectrum, the bremsstrahlung conversion target served as appropriate filter. The experiment on double-sided electron processing was repeated also with a larger number of items and dosimeters applied; the shape of the resulting frequency distribution of dose is still quite close to the normal distribution (Fig. 5) and only at the highdose side the practical upper limit for dose is encountered around 2.5 kGy.

TABLE 4. CHARACTERISTICS OF DOSE DISTRIBUTION IN DOUBLE-SIDED ELECTRON PROCESSING AND SINGLE-SIDED BREMSSTRAHLUNG PROCESSING OF PISTACHIO (dose in kGy)

	mean	std.	Min	max	max/min
10 MeV electrons	1.31	0.261	0.995	1.79	1.80
(repetition/normalized)	1.62	0.227	1.00	2.50	2.50
5 MeV X rays	0.793	0.121	0.661	1.04	1.57
10 MeV X rays	0.818	0.123	0.653	1.03	1.58

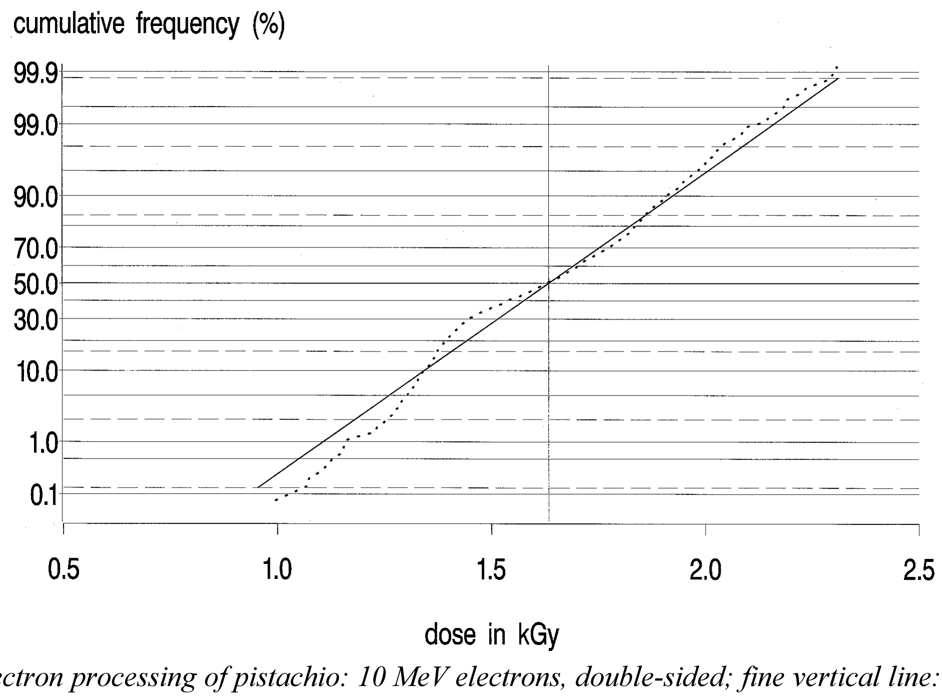

FIG. 5. Electron processing of pistachio: 10 MeV electrons, double-sided; fine vertical line: mean value (experiment with 23 repetitions and 1400 individual dosimeter readings.)

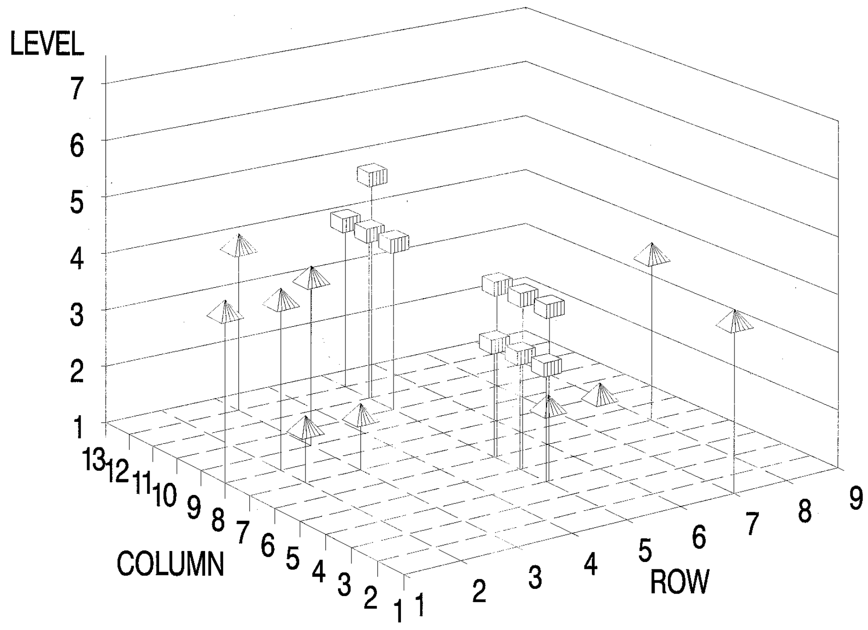

FIG. 6. Position of the 10 largest and of the 10 smallest dose values for 10 MeV electron processing (double-sided) for the repetition experiment on pistachio (dose mapping by 819 individual dosimeters): level of occurrence by row and column.

The experiment on double-sided electron irradiation was repeated in a set of 23 runs and a dose mapping run in the same geometry as described before. 6 layers of pistachio pouches allowed for 7 levels of dosimeter placement. Each layer consisted of two rows and 4 positions (resp. 9 rows and 13 positions in the mapping experiment) for the dosimeters. The dose mapping study revealed (Fig. 6) that the maximum dose mainly occurred in the centre of the boxes and along the direction of conveyor movement; the minimum doses also occurred mainly inside the box and not at surfaces. This effect was not caused by any inhomogeneity of dose distribution in scanning direction, but is due to the geometrical arrangement of the pouches in the boxes: through the centre of the pouches the beam passes most mass and the minimum dose is likely to occur in the middle of its trace; between the two rows of pouches the edges overlap, less mass is encountered and the maximum dose is due to the two-sided overlay of the largely unattenuated beam. The repetition run on electron processing was at a higher nominal dose and, therefore, for direct comparison normalized to equal minimum dose of 1.0 kGy (Table 4). The fluctuations of the overall dose distribution (standard error 14%) can be compared with the results from the reference dosimetry (dosimeters on each box in the centre facing the source and replaced when turning the boxes for the second pass); the mean reading of the control dosimeters was 1.052 kGy (standard error 2.2%), the observed lowest dose value was 1.00 kGy, and all variations of dose were in the range above the required minimum dose. Consequently, for this particular irradiation geometry it may be concluded that the surface dose of a single pass is closely related to the minimum dose and about 5% above that value. In the case of electron processing the surface dose is a particularly suited quantity for reference as it is directly linked to machine setting, ie mean current, scanning width and transport velocity. The higher max/min ratio for the repetition reflects that the geometry of pouches in the cardboard boxes during the 23 repetitions fluctuated considerably. In order to save costs the pistachio boxes were reused several times; the dosimeters had to be inserted and retrieved for which purpose the stacked pouches had to be removed from the box and laid in again.

3.2.4. General aspects and summary

Under this CRP from the very beginning and with respect to its duration of only five years, it was agreed that it would not be possible to collect a body of data on dose distribution occurring under a variety of practical conditions and covering all relevant geometries large enough to serve as a foundation of sound inter and extrapolation of frequency distributions of dose and of reliable estimation of the parameters of such frequency distributions from a limited set of dose measurements at some reference positions. The resolution of this problem, however, was seen as a prerequisite for the use of 'label dosimeters' in quarantine inspection of produce disinfested by ionizing radiation. The data reported in this contribution and by other participants of the CRP show at least that such approach is possible in principle. The pistachio experiment reported above shows that for special geometries here double-sided electron irradiation of boxes with a height in beam direction equivalent to electron penetration and where the dose value at the reference position is rather close to the dose value at the expected position of the minimum dose such conclusions are possible and allowable. In many practical situations an accessible position to attach some label dosimeter would not be a suitable and reliable reference position for dose measurements. Dose fluctuations on most positions at the surface of containers for the majority of irradiation geometries might be too large to allow for reliable estimation of the characteristic parameters of the frequency distributions of dose belonging to the respective setups. This would be especially true for many sided irradiation when the reference dosimeter can not be retrieved between single passes through the radiation field.

4. PART B: CHARACTERIZATION OF A LABEL DOSIMETER

In principle, there are two types of label dosimeters: The first type could consist of a barcode the optical visibility of which changes with dose and, thus, could give the dose reading; it also could be a threshold indicator for the required minimum (or reference) dose. The second type could be any conventional dosimeter which can be read by a suitable meter while still on its place on the container. Especially dosimetry films which are already in common use could be read in reflectance mode. Also such threshold dosimeters could be judged upon visual appearance.

Label indicators as used by the radiation processing industry for visual aid in inventory control are often thought to measure dose or to ensure a certain minimum dose. However, such labels do not have any metrological property. Instead, stamping every item emerging from the irradiation chamber would have the same documentary character when linked beyond all doubt to the process records of the facility. Furthermore such indicators typically are attached to the product load at positions on the outer surface of the goods and the relationship between dose at such reference position and dose at the expected position of the minimum dose is not established generally. The problem remains to establish such a link which must also be acceptable to control authorities.

Label dosimeters, on the contrary, would be instruments measuring the dose or 'indicating' that a given threshold value of dose was surpassed at the position where the 'label' resides. The metrological properties of such devices with regard to the nominal dose threshold and the reproducibility of the indication was to be studied; environmental factors also should not be neglected. Insect eradication and quarantine regulations might become a challenging application of such 'label' devices. Especially in the light of the statistical approach taken above, the value of such devices remains disputable, even after they have been proven to be real dosimeters: Securing a given dose value at an easily accessible position outside a consignment not necessarily ensures at the same time that a minimum dose throughout the product load has been met even at positions not accessible for verification. To ensure that, other measures are indispensable.

4.1. Materials and methods

"STERIN 70", "STERIN 125", and "STERIN 300" 'indicators' were used, which were made available to members of this CRP by its manufacturer (International Speciality Products (ISP) Inc., Wayne NJ, USA; similar indicators with the trade name "RADSURE" are used to a certain extent by the blood banking community). Type '125' came with a clear release film on top of each which could be peeled to expose an adhesive and to attach the indicator on the inside of a clear container; all measurements were taken through that film without peeling it. Types '70' and '300' came without such release film on the surface.

STERIN 'indicators' have a sensitive red field which darkens to black with increasing dose. In this read field the text 'NOT' in black on the red background is seen for the unirradiated indicator; as the red background darkens at a certain dose level the 'NOT' can no longer be read. The nominal dose (ie 70, 125 or 300 Gy) is also imprinted on the label indicator. Thus, the inscription initially is 'NOT irradiated at nnn Gy' which changes to 'ÛÛÛ irradiated at nnn Gy' around the threshold dose.

Colour measurements were taken with a tristimulus colormeter (Minolta Chroma Meter II 'reflectance') in CIELab coordinates. Lab coordinates were preferred as these most closely reflect human perception of equal colour differences. Furthermore, Lab coordinates allow for illumination and viewing conditions. At the same time this property is also a limitation with regard to comparison between laboratories: illumination and viewing conditions are difficult to reproduce. The tristimulus reader used here had internal conversion of measurements using predetermined factors. The conversion to other colour coordinates as YXZ or Yxy systems is not straight forward and several sets of conversion equations have been published. Despite this deficiency the results reported in the CIELab coordinates are comparable between participating laboratories and the results are in general agreement.

Subjective judgements were taken by an untrained panel of 25 participants each asked to rank a set of sample indicators irradiated at several dose levels or to discriminate the odd sample of three indicators irradiated at two dose levels (one of the three being a replica of the two dose value presented). For ranking, sets of six label dosimeters each were presented to the judges in random order; each dosimeter of the six treated with 60, 80, 90, 100, 110 and 120 % of the respective nominal dose; the judges were requested to arrange the labels in order of blackening of the 'NOT'. In a series of five triangle tests two labels irradiated at 120 % of the respective nominal dose together with an odd sample treated at 60–110 % of respective nominal dose were presented and the test was to identify the odd sample. The odd sample of the triangle sets was always presented at random position in the triplet. The indicators were labelled by three digit random numbers in both tests in order to prevent prejudice from sample naming; the triangle sets were disclosed to the judges in a gambling manner in order to prevent recognition of the two (repeating) 120 % reference samples and their codes; this was repeated for the five sets. Testing for unbiased judgement (asking individuals who had never seen a label before to read the complete text on the label and subsequent evaluation of the answers whether the 'NOT' was recognized) was not considered feasible because of burden of such extensive work. Results of the research into validation by subjective and objective means for such labels was already reported elsewhere; only a condensed summary is given below.

4.2. Results and discussion

Disputable remains the value of such label dosimeters while they have been proven suitable and reliable by the studies during this CRP. Securing a given dose at an easily accessible position outside a consignment not necessarily ensures that the minimum dose is met throughout the product load and even at positions not accessible for verification.

All label indicators showed the obvious effect: the 'NOT' became invisible when the red field turned black after receiving the indicated threshold dose or more. This effect was followed up by colour measurements in reflectance mode and by visual judgments using a test panel. The colour change in the abplane of the CIEcolour space occurred on a straight line from higher chromaticity (upperright corner) with increasing dose to the neutral chromaticity point at the coordinates' origin for all three indicator types (Fig. 7). All types of indicators were irradiated with gamma rays and electrons (the latter not for "STERIN 70"). The length of the distance passed with increasing dose from the respective starting point of the unirradiated samples is used in the following graphs; the course of CIElightness (brightness) is also given (Figs 8–10). It can be seen for all three indicator types that distance and lightness approach their asymptotic values around the respective nominal dose value; both parameters separately may be used for discrimination. Therefore lightness is used for comparison with instrumental reading by a prototype 'dose indicator reader'.

The study also included environmental factors such as temperature and UV light; humidity effects were not investigated. Storage of STERIN 'indicators' at 25 and 40 °C as well as under UV light (366 nm) did not cause significant changes even over 100 h storage/exposure; the variations are within the reproducibility of the colour readings.

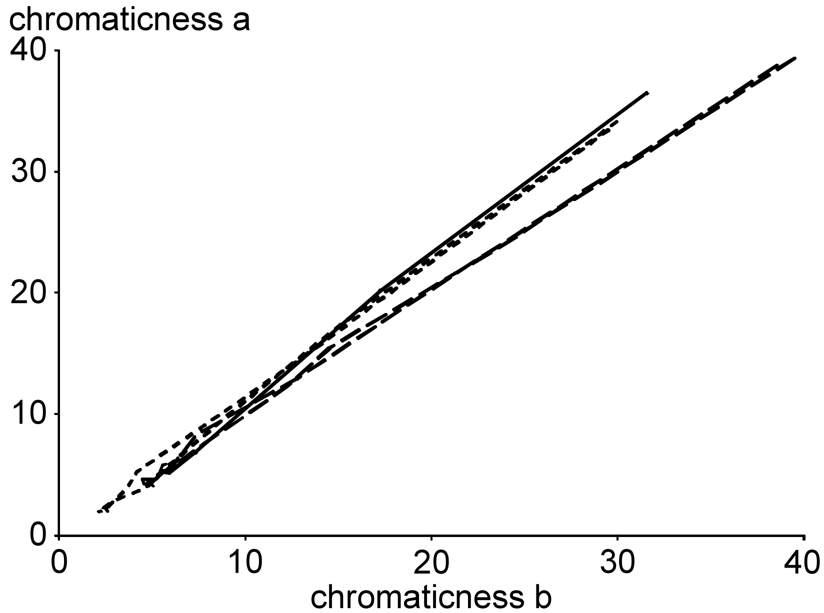

FIG. 7. Colour change for dose indicators, in CIEa and b chromaticity coordinates. increasing dose from upperright corner (red/yellow) to neutral at origin of the coordinate system (chromaticity direction to blue/green); indicator thresholds 70 Gy and 125 Gy (solid lines) and 300 Gy (dashed lines); electron and gamma irradiation (electron not measured for 70 Gy indicators) are given in identical line types as they fall close to each other.

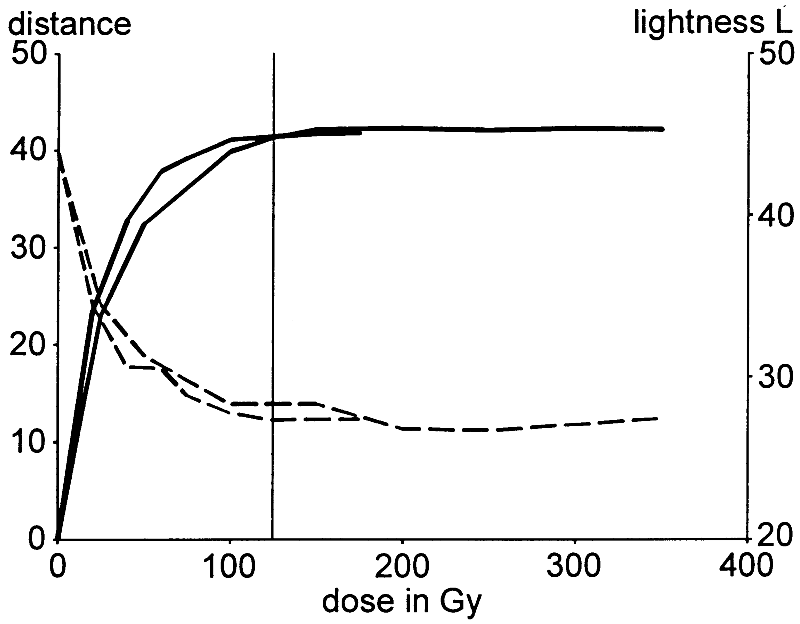

FIG. 8. As a function of dose, the arc length (see FIG. 7) traversed in chromaticity space (left scale, solid line) as well as the lightness in CIEL dimension (right scale, dashed line); treatment by cobalt-60 gamma rays; indicator threshold 70 Gy (fine vertical line).

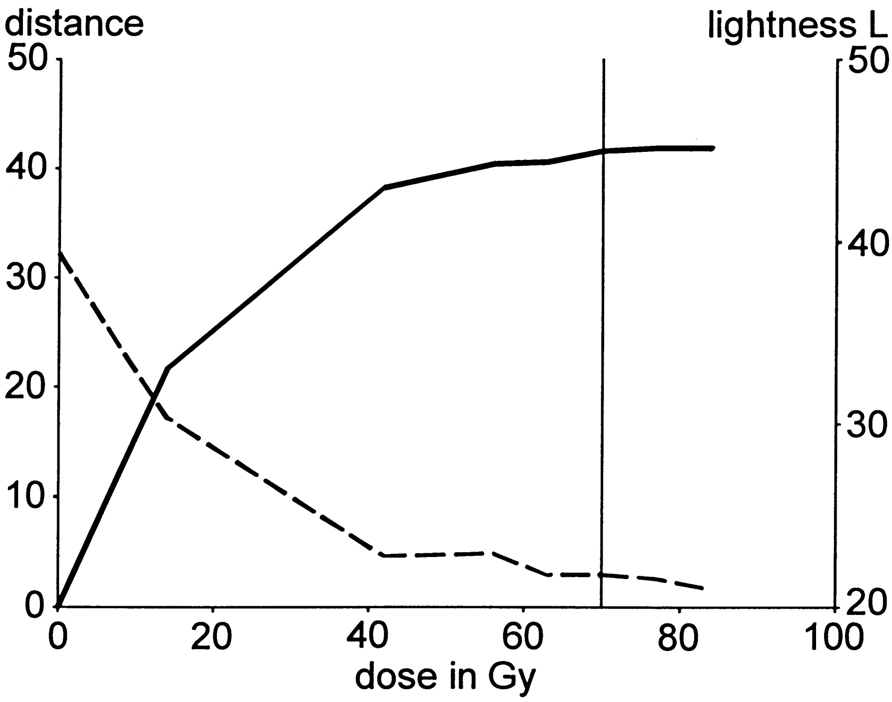

FIG. 9. As a function of dose, the arc length (see FIG. 7) traversed in chromaticity space (left scale, solid line) as well as the lightness in CIEL dimension (right scale, dashed line); treatment by cobalt-60 gamma rays and 10 MeV electrons, hence, two set of solid and dashed lines; indicator threshold 125 Gy (fine vertical line).

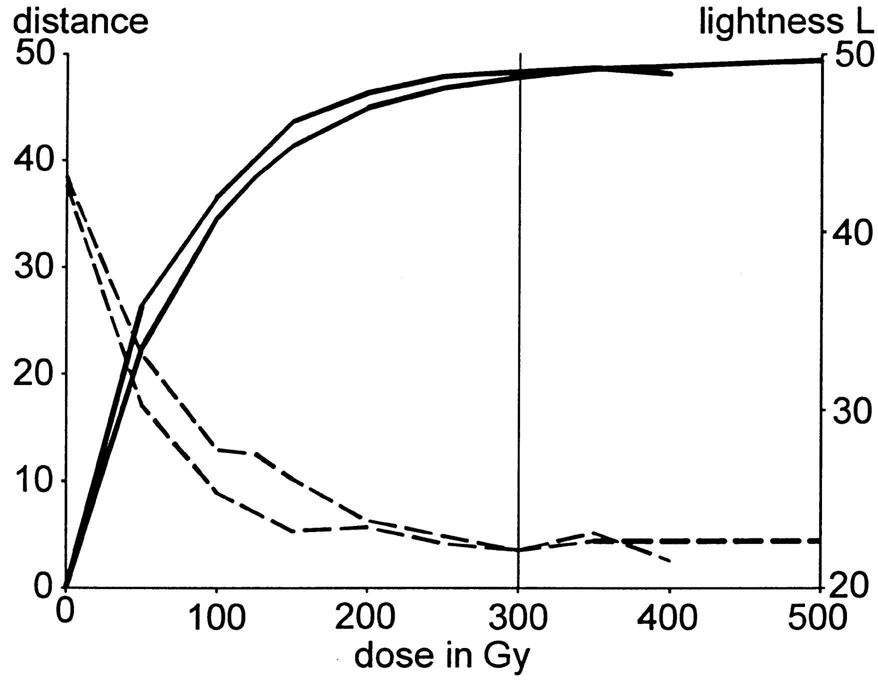

FIG. 10. As a function of dose, the arc length (see FIG. 7) traversed in chromaticity space (left scale, solid line) as well as the lightness in CIEL dimension (right scale, dashed line); treatment by cobalt-60 gamma rays and 10 MeV electrons, hence, two set of solid and dashed lines; indicator threshold 300 Gy (fine vertical line).

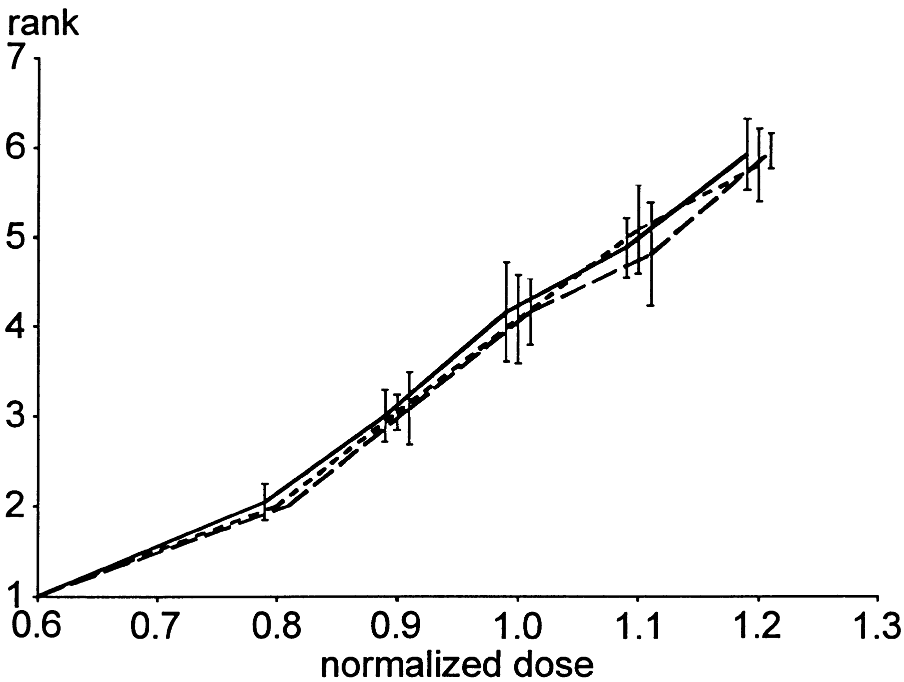

FIG. 11. Average ranks (25 judges); indicator threshold 70 Gy (solid line), 125 Gy (dotted line), 300 Gy (dashed line); 70 Gy and 300 Gy displaced to left and right, respectively, for better visibility; standard deviation indicated; dose normalized to indicator nominal dose of 70, 125 and 300 Gy.

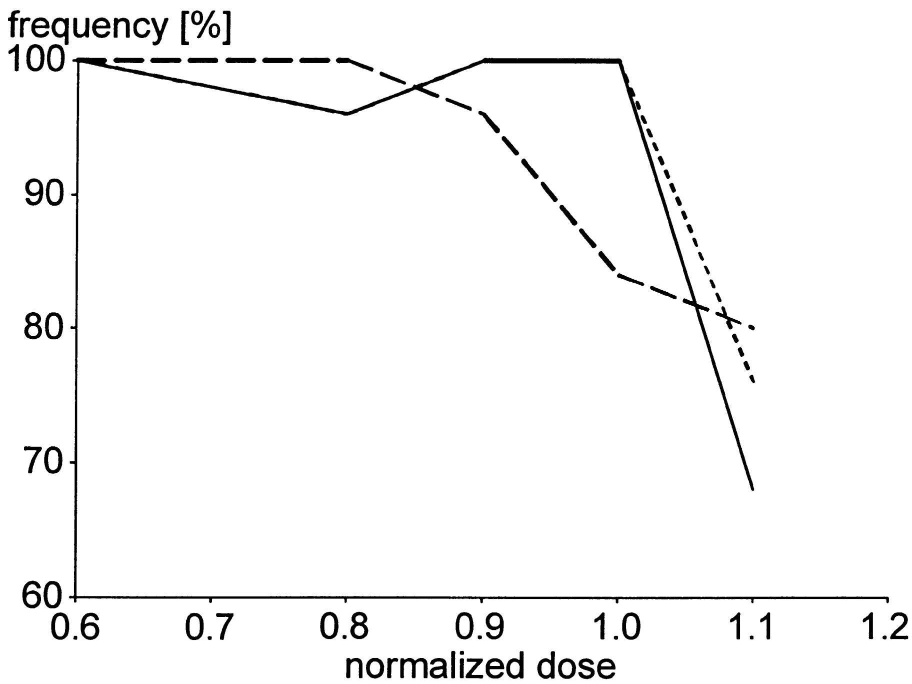

FIG. 12. Frequency of correct recognition of the odd sample (25 judges); line types for indicator nominal dose same as in FIG. 11; dose normalized to indicator nominal dose of 70, 125 and 300 Gy.

The results of the ranking test (Fig. 11) make obvious that the judges could discriminate and arrange the samples in a very reproducible manner; only directly neighbouring dose steps were sometimes arranged incorrectly which results in the rather small standard deviations for rank averages. Testing for least significant distances (á = 0.01, Duncan test) revealed that all dose steps are discriminated by their mean ranks for the three indicator types, respectively.

In triangle tests the guess probability is 1/3; if more than 17 judges out of 25 find the correct answer this is highly significant (á=0.01). A large group of judges was able to discriminate still 110 % against 120 % nominal dose for each type of labels (Fig. 12). For the labels with nominal dose of 300 Gy the decision dose is not as sharply defined as for the other two nominal doses. Practically, however, this implies that for type '300' only one judge out of 25 could not see for 90 % nominal dose the difference from the black reference ones; at 100 and 110 % of nominal dose this group increases to four to five judges among 25. All judges could see the 'NOT' being blackened away, but the shade of that black was still different enough between samples and only allowed for discrimination. However, not the shade of black but the readability of the 'NOT' is the validation criterion. It should be noted that the judges of the testpanel always tried to use any additional information (eg glossiness of label surface) for better discrimination and for improved performance in the sense of possibly expected answers.

5. PART C: DEVELOPMENT OF A LABEL DOSIMETER READER
(in cooperation with B. Bauer)

By subjective and objective judgements it could be shown that STERIN labels have suitable metrological properties; other participants of this CRP also contributed. Consequently, the idea was developed to design a simple, hand-held reader which could replace subjective judgements and which could be as reliable as a thorough spectrophotometric or colorimetric analysis. The principle approach was to replace prism, grid or filters as wavelength analysing instruments by diode illumination at suitable wavelength; 615 nm was chosen for the experimental setup (Fig. 13). The reflected light is then measured by a standard photodiode. Split glassfibre optics is used to illuminate and measure at the same spot. Simultaneously the type of the indicator imprinted close to the radiation sensitive window is read and all information are processed in some dedicated computer; the respective calibration function for several types of indicators may be selected. The whole instrument (Fig. 14) can be converted into miniature form or a hand-held device (Fig. 15). The indicator might be inserted in a reading slot or the reader might be put on top of the indicator still on its place on a carrier load.

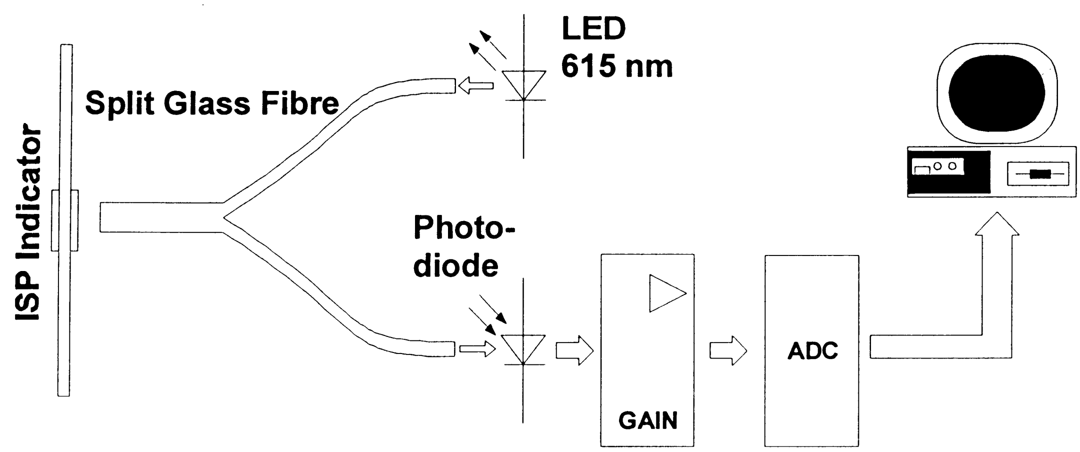

FIG. 13. Schematic diagram of the experimental setup for instrumental indicator readout.

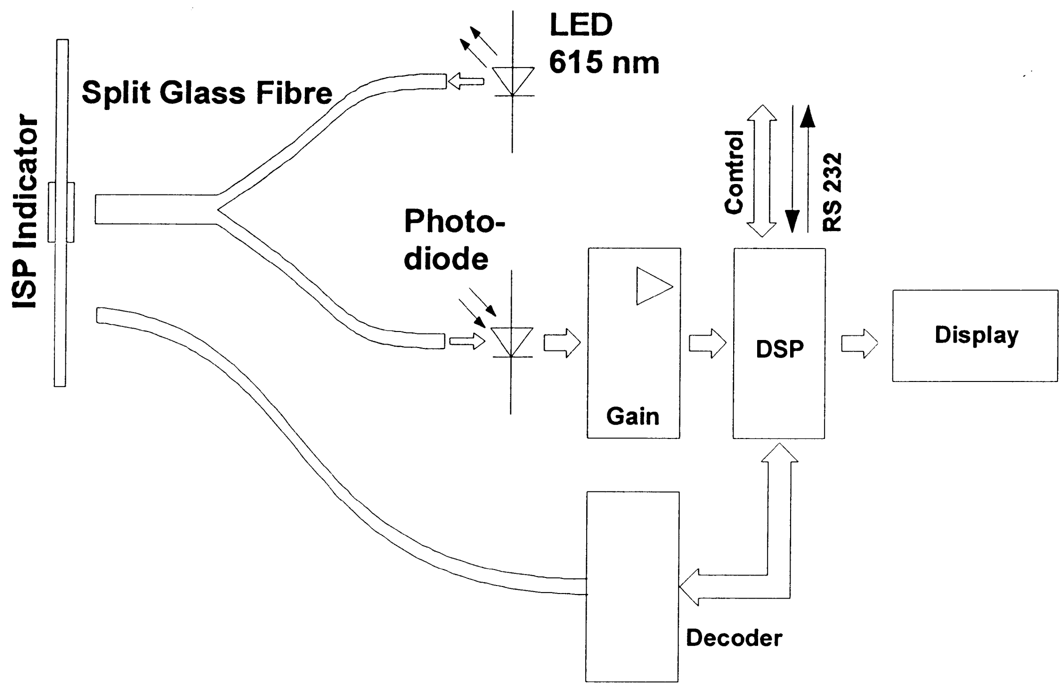

FIG. 14. Schematic diagram of the prototype reader for label dose indicators.

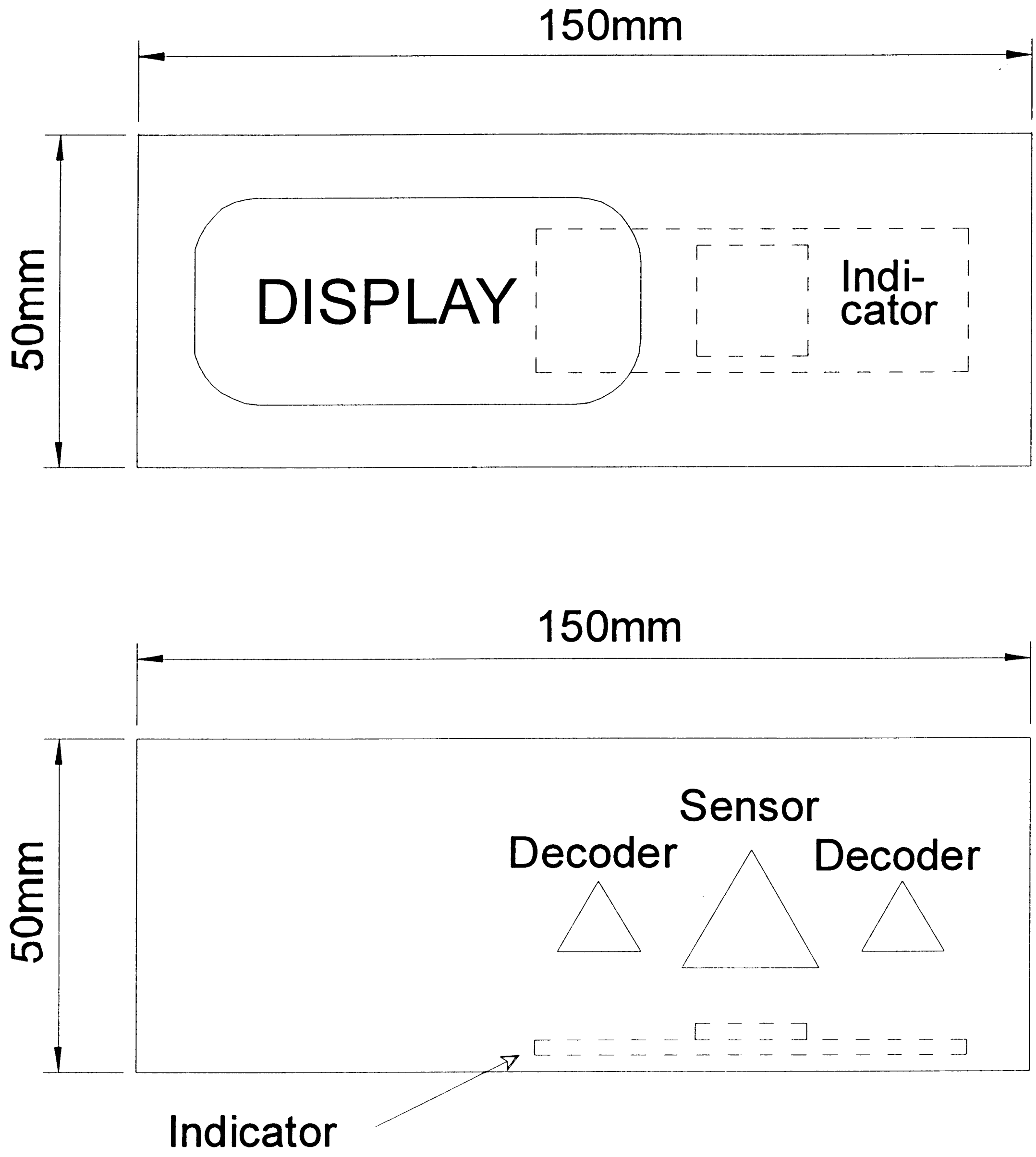

FIG. 15. Design of a miniaturized, hand-held label indicator reader for routine applications.

The experimental set up was used for reading the STERIN 125 and 300 indicators; results were compared to colorimeter readings of CIEL brightness. All data are normalized from their respective range to a common range of 1 to 0 for comparison (Figs 16 and 17); agreement between methods and for repetition is reasonable. The use of the proposed miniature reader (Fig. 15) is considered promising. Such equipment would need to identify the type of the 'label dosimeter', apply an appropriate decision function derived from the several response curves of the given sample and adjust for the required reliability of the judgement. This might become difficult as the comparison (Figs 16 and 17) of photodiode readings shows: the general trend is clear but some indicators (here STERIN 300) passes the threshold (here 300 Gy) by far too early (here 125 Gy). Much more work would be required to make such 'label dosimeters' finally acceptable for application in quarantine inspection of fruit disinfested by ionizing radiation.

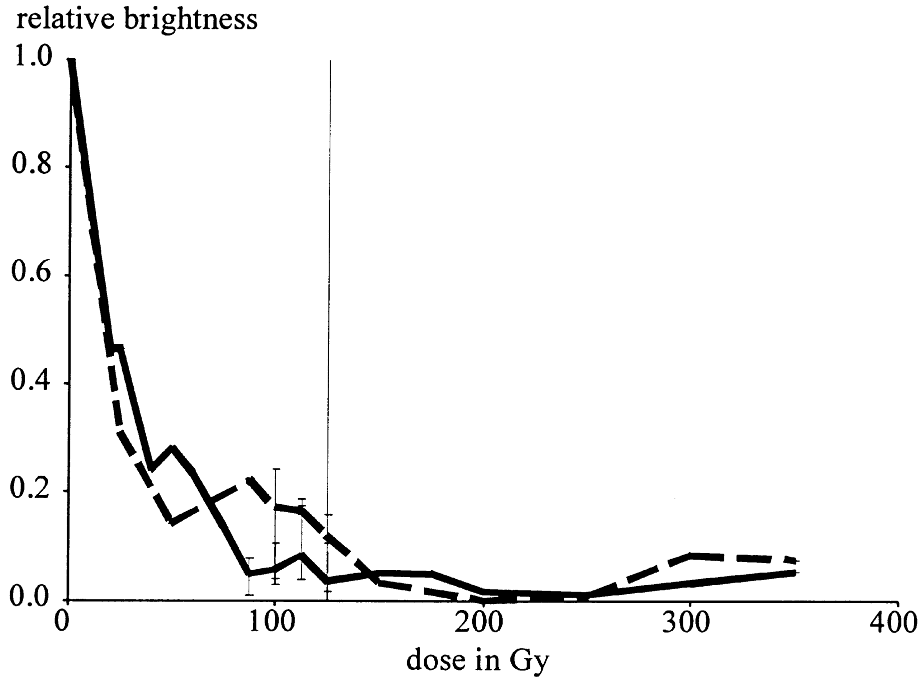

FIG. 16. Comparison of CIEL brightness (solid line) and photodiode (prototype reader, dashed line) signal for indicator nominal dose of 125 Gy; respective raw data normalized to range 0 1; fine vertical line indicator threshold; max/min of repeated measurements indicated.

FIG. 17. Comparison of CIEL brightness (solid line) and photodiode (prototype reader, dashed line) signal for indicator nominal dose of 300 Gy; respective raw data normalized to range 0 1; fine vertical line indicator threshold; max/min of repeated measurements indicated.

6. SUMMARY

The availability of the first commercial dose level indicator prompted attempts to verify radiation absorbed dose to items under quarantine control (eg for insect disinfestation) by some indicator attached to this item. The fundamental problem, however, would be the question whether a dose read at a certain reference position can be linked in a reliable and clearcut way to the respective minimum dose. Studies into variability of dose distributions under model situations reflecting commercial practices revealed the difficulty of such approach. Until now, only in situations where the position of the minimum dose is accessible and the indicator can be attached to this position a valuable dose judgement is possible. In many commercial situations the minimum position is inside the package or container and not accessible. Therefore, still more research into the statistical nature of dose distributions and into the link between measured values at reference positions to the parameters of such dose distributions is indispensable.

Samples of the new commercial dose level indicators were tested for their metrological properties. The devices are suitable for the intended purpose and the subjective judgement whether the threshold dose was surpassed is possible in a reliable manner. It should not be overlooked that the ranking and triangle tests reported here are overcritical compared to the routine situation of their intended application. Also instrumental measurements of the colour and brightness change of the indicator were executed. The subjective judgements are completely backed by the instrumental results. Consequently, a prototype reader was developed; first tests were successful.

ACKNOWLEDGEMENTS

The technical assistant Michael Knörr with support by the students Christiane Soika, Karin Weiss, Britta Mager conducted the large number of dosimeter readings and especially the subjective judgement on label dose indicators; Michael Knörr finally converted the results in fine graphical presentations; the personnel of the contract irradiator (Gammaster Allershausen) cooperated in an outstanding manner and contributed their dose readings. Gamma processing was done free of costs; ISP Wayne provided free of cost sets of prototype label indicators to all participants and extra samples for the studies reported here. This work and contribution is gratefully acknowledged; thanks go also to IAEA for accepting this research proposal without which the initiative for the reported work would have been missing and this research in principle necessary would not have been conducted.

REFERENCES

[1] Anon., Papaya, Carambola, and Litchi From Hawaii (7 CFR Part 318 [Proposed Rules]), Federal Register 61 (1996), No. 142, 38108–38114].

[2] Chadwick, K.H., D. Ehlermann and W.L. McLaughlin, Manual on Food Irradiation Dosimetry, Technical Reports Series No. 178, IAEA, Vienna (1977).

[3] Ehlermann, D., The use of various dosimeters for the measurement of random fluctuations of the dose distribution in commercial scale food irradiation, in: Dosimetry in Agriculture, Industry, Biology and Medicine, IAEA, Vienna (1973) 77–83.

[4] Ehlermann, D.A.E., Process Control and Dosimetry Applied to Establish a Relation Between Reference Dose Measurements and Actual Dose Distribution, Second Research Coordination Meeting on "Standardized Methods to Verify Absorbed Dose in Fresh and Dried Fruits, Tree Nuts in Trade", Federal Research Centre for Nutrition, Karlsruhe, Germany, August 1996.

[5] Ehlermann, D.A.E., Label Dosimetry as a Means to Verify Dose, Second Research Coordination Meeting on "Standardized Methods to Verify Absorbed Dose in Fresh and Dried Fruits, Tree Nuts in Trade", Federal Research Centre for Nutrition, Karlsruhe, Germany, August 1996.
[6] Ehlermann, D.A.E., Validation of a Label Dosimeter for Food Irradiation Applications by Subjective and Objective Means, Appl. Radiat. Isot. **48** (1997) 1197–1201.
[7] Razem, D., Dosimetric Performance of and Environmental Effects on STERIN Irradiation Indicator Labels, Radiat. Phys. Chem. **49** (1997) 491–495.

DETERMINATION OF MAXIMUM/MINIMUM RATIO OF ABSORBED DOSE OF DRIED FIGS

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Abstract. In the framework of FAO/IAEA project, the ECB dosimeter and STERIN-125 and STERIN-300 dosimeters have been used for dose measuremennt in the dried figs packs. They were irradiated in our Gamma Irradiation Plant and were given 6 kGy dose. It was observed that all Sterin label dose indicators became very dark after a 6 kGy dose and the absorbance could not be measured with UV spectrophotometer. Therefore these label dose indicators were separetely irradiated between 10–700 Gy doses by gamma rays to establish the dose sensitive curve of these indicators. After the irradiation of ECB dosimeter which is located in dried fig packs, we found the Dose Uniformity Ratio as 1.4 according to bulk density of 0.62 gr/cc.

1. INTRODUCTION

About 55,000 mTon of dried figs are produced annually in Turkey (about 60% of total world production) with 30,000 mTon or more being exported (over 70% of total world export). This compares with 250,000 mTon/year of fresh figs for the domestic market and also for export, mainly to Europe and the Middle East.

Dried fig packs have a bulk density of from 0.6 to 0.8 g/cc which happens to be the product density at which the irradiator operates most efficiently according to theoritical calculations that also show a projected max/min. dose ratio of 1.4 for dried figs in standard pack, which contains 60 small packs of 1 kg each. Studies under contract No. 7779/RB were found as experimentally the same dose uniformity ratio according to bulk density of 0.6 g/cc figs.

STERIN-125 and STERIN-300 label dose indicators were also used during the irradiation of figs.

2. GAMMA IRRADIATION PLANT

The Gamma Irradiation Plant in Ankara Nuclear Research Center has been working since earlier of 1993. The Hungarian designed and built irradiator is of the 4pass product overlap type, with a full cell load of 52 tote boxes (45 × 45 × 44 cm) two per carrier which shift positions vertically during cycling. There are 24 carrier dwell positions, 6 per row for a total of 48 totes, plus an exit and an entry carrier for a total of 52 totes in the cell at any given time during continuous and batch mode operation with an initial loading of 100 kCi of Cobalt-60 (15.01.1993 and now 52 kCi and highest dose rate 3.8 kGy/hr) Medical disposibles (syringes, catheters, infusion sets, etc.) are irradiated with the standard 25 kGy minimum

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sterilization dose. At a typical bulk density of 0.17 gr/cc the plant sterilizes 1250–1500 m^3/year.

The Ethanol Chlorobenzene (ECBRO) dosimeter is used [1] with the Fricke calibration reference standard dose mapping and routine dosimetry.

3. ETHANOL CHLOROBENZENE DOSIMETER

A disadvantage of Fricke dosimetry is its unsuitability to measure doses above 400 Gy. The accuracy of alcoholic chlorobenzene dosimetry does not reach that of the Fricke method, but it is much more applicable for routine measurements owing to its simplicity, relative insensitivity to impurities and wide dose range of measurements.

The dosimeter [2] has a high thermal stability and resistance to oxidation and the yield of radicals in irradiated chlorobenzene depends on concentrations. Ethyl alcohol, aceton and water are introduced into the system to stabilize the chlorine atoms produced in the radical process and their transformation into the chloride ions. The alcohol also inhibits the chain reactions of oxidation and serves as a good solvent for hydrochloride acid. Evaluation can be by titration or oscillometric measurement. Oscillometry belongs to the group of conductivity measurements and is carried out by means of the high frequency alternating current. It has a linear relation hydrochloric acid production with absorbed dose in the range of 0.5–400 kGy with the upper limit being governed by the dose rate.

3.1. ECB dosimeter characteristics

The properties of this chemical dosimeter can be observed:

- The clorine ion formed from irradiated clorobenzene is in the form of hydrochloric acid, it is stable in solution under irradiation even at a concentration of 0.5 mol/L.
- Clorobenzene solution can be saved for long time.
- This solution and dosimeters can be prepared easily.
- The initial value of G ($Cl^)$ at a given chlorobenzene concentration (4–40vol%) is independent of the dose in the dose range 200 Gy–$4*10^5$ Gy.
- The accurracy of determination remains unchanged between the dose rates 0.045 and 25 Gy/s.
- This dosimeter is temperature independent between 20^0C–90^0C.
- Correct dose data are obtained if the irradiated ampoules are kept in darkness for several years and evaluated afterwards.
- Chlorobenzene is thermally stable and resistant to oxidation.
- A comparison of the titration data with those obtained by oscillometry shows that the deviation exceeds % +5.

In preparation, 240 ml of chlorobenzene and 40 ml of distilled water are measured into 1000 ml flask, 0.4 ml of aceton and 0.4 ml of benzene are added and the flask filled with water free alcohol up to the mark and used for fig irradiation. This irradiated ECB dosimeter was measured on a OK302 type oscillator.

3.2. Principles of RO(readout) instruments

The conduction of ECB solution is changed by the alteration of the amount of ions present in the solution according to Kohlraush's Law. This phenomenon is observed by measuring the conductivity by high frequency oscillometry. There is no a connection between the solution and electrodes in high frequency conductivity measurements. The advantage of the method is that with the help of oscillometry the conductivity measurements can be carried out in a completely closed system such as in sealed ampoules. The dose information can be stored.

3.3. Calibration of chlorobenzene dosimeter

The calibration [3] can be carried out by measuring the conductance of a solution irradiated with a known dose. If the dose is unknown, the titration can be applied to determine the dose. The evaluation is very simple by using calibration curve or chart, or power regression analysis programme. We used 3.3 kGy, 4.5 kGy, 8.7 kGy, 14.1 kGy and 28.2 kGy in 2 cm^3 sealed ECB ampoules [4] for calibration which were sent to us from Institute of Radioisotopes of Hungary. The sensitivity of instrument should be often checked by calibration of ECB ampoules. ECB and the reference dosimeter ampoules should be held at the same temperature for one hour before the evaluation with readout (see Fig. 1).

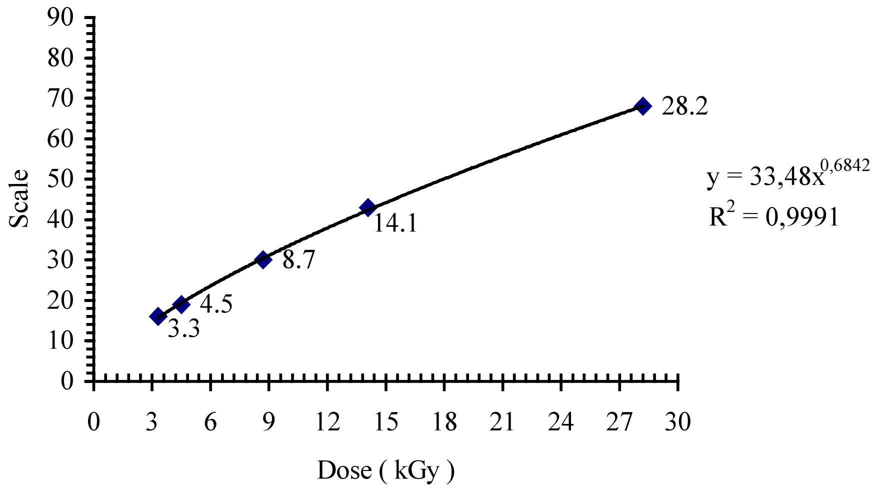

FIG.1. Calibration curve of chlorobenzene dosimeters.

3.4. Evaluation of irradiated ECB dosimeter

We used 27 ECB ampoules with STERIN dose indicator during irradiation of fig boxes that have been located [5] as in Figure 2. Irradiated ECB dosimeter were evaluated by RO instrument that is made in Hungary and all evaluation of ECB was made in the room temperature which was given around 6 kGy in Gamma Irradiation Plant. It was found the minimum dose as 5.83 kGy in the middle and maximum dose as 8.14 kGy in 0.62 g/cc bulk density of dried fig packs. According to result of evaluation of ECB, *Dose Uniformity Ratio* (D_{max}/D_{min}) was found to be 1.4 for 0.62 g/cc bulk density of dried fig packs (see in Table 1).

This ratio comfirmed that the efficiency of gamma irradiation Plant is the same theoretically [6] and experimentally for 0.6 g/cc bulk density of products.

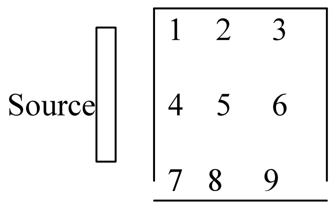

FIG 2. The position of the ECB dosimeters in dried fig pack.

TABLE 1. DOSE DİSTRİBUTİON İN DRİED FİG PACK (kGy) İRRADİATED AT GAMMA IRRADİATİON PLANT OF ANKARA NUCLEAR RESEARCH AND TRAİNİNG CENTER (ANAEM)

ECB(kGy)	1	2	3	4	5	6	7	8	9
TOP	7.27	6.23	7.92	7.49	5.83	7.92	7.49	6.44	8.14
MIDDLE	6.85	6.23	7.06	6.64	5.83	7.06	7.27	6.23	7.49
BOTTOM	6.67	5.83	6.64	6.74	5.86	6.64	7.26	5.95	7.64

4. STERIN-125 AND 300 Gy RADIATION DOSE INDICATORS

STERIN minimum radiation dose indicator is the new product of International Specialty Procucts ISP Dosimeter Division of GAF Industries, Wayne, New Jersey, USA. This label dose indicator has a radiation sensitive window which display the visual message “NOT IRRADIATED”. After irradiation the sensitive window becomes opaque to obscure the word “NOT” and change the visual message to “IRRADIATED”. The STERIN indicator can be easily attached to any product container and the sensitive dose range between 10–500 Gy. In this dose intervals, it must be kept away from exposure of direct light such as ultraviolet,visible and infrared.

4.1. Test of STERIN-125 and -300 Gy label dose indicators in gamma ırradiation plant

STERIN-125 and -300 were irradiated in Gamma Irradiation Plant with Co^{60} plate source of 100 kCi (15.01.1993). Dose rate of irradiation was 338 Gy/h. The dose employed were 10, 50, 70, 100, 125, 150, 200, 300 Gy for STERIN-125, and 10, 50, 100, 150, 200, 300, 500 and 700 Gy for STERIN-300 and the response curve of both STERIN dose indicators were determined (see Figs 3 and 4). After irradiation, these plastics were detached from the paper label and cleaned with ethyl acetate to remove the adhesive glue. We used Ati Unicam UV4/V is spectrophotometer for measurement of the STERIN indicators and thickness measurement was found as 0.576 ± 0.002 mm for 125 Gy and 0.375 ± 0.002 mm for 300 Gy STERIN indicators. The maximum peak of the optical density spectrum (ΔA/l)for 125 and 300 Gy indicators was defined at 674 nm. It was seen that the label appeared completely dark when the irradiation dose was 125 Gy and above. Similarly, the word NOT was completely dark when irradiation dose was 300 Gy for STERIN-300 indicators.

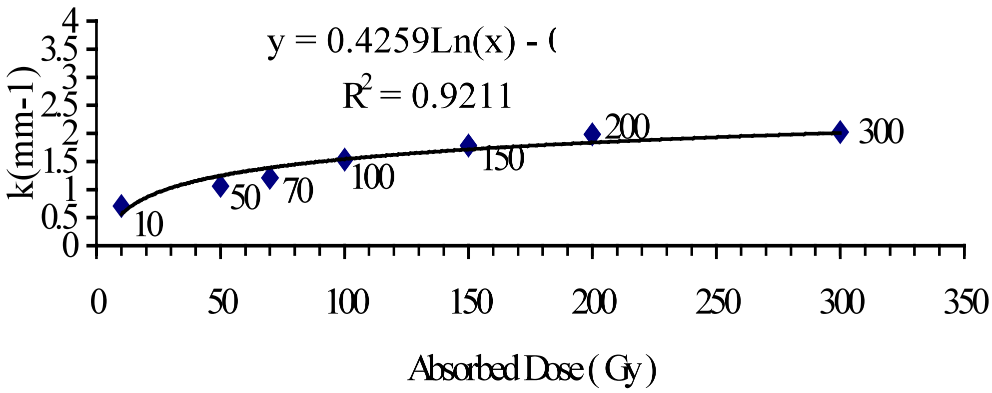

FIG.3. Response curve of STERIN-125 Gy dose indicator at 674 nm irradiated by Co^{60}.

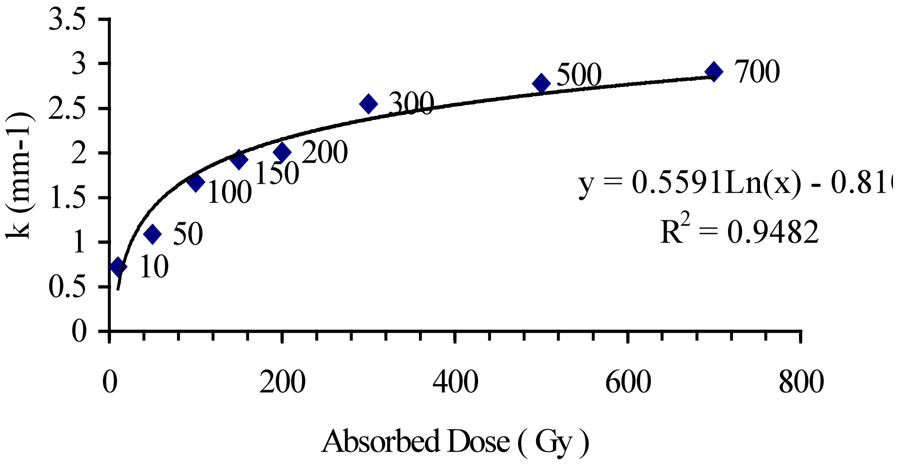

FIG. 4. Response curve of STERIN-300 Gy dose indicator at 674 nm irradiated by Co^{60}.

In addition STERIN-125 and 300 Gy label dose indicators were used in containers of figs irradiated to a dose of 6 kGy. It was seen that all indicators were very dark and the absorbance could not be read in a UV spectrophotometer. Therefore, we can't use these dose indicators for max/min dose uniformity ratio in bulk density of fig products. STERIN label indicators can be used in Gamma Irradiation Plant as a label dose indicator for different kind of products such as spice and dried foods for the commercial scale.

5. FINAL REMARKS

Dried fruits such as as figs, grapes, apricots and hazelnuts are very important products for the economy of Turkey. Therefore radiation processing of these items by sure will definitely be part of future discussions in our country. In industrial scale, the STERIN-125 and

300 Gy label dose indicators can be recommended for radiation proccessing as label dose indicator and can be used for large scale irradiation of food products.

ACKNOWLEDGEMENTS

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REFERENCES

[1] RAZEM,D., DVORNİK,I., "Application of the ethanolchlorobenzene dosimeter to electron beam and gamma radiation dosimetry. II.Cobalt-60 gamma rays", Dosimetry in Agriculture, Industry, Biology and Medicine (Proc. Symp. Vienna, 1972) IAEA, Vienna (1973) 405.

[2] STENGER,V., TORDAY,ZS., HORVATH,I, "Long term experience in using the ethanaol chlorobenzene dosimeter system ." Proc. International Symposium on High Dose Dosimetry for Radiation Processing, IAEA, Vienna (1990).

[3] STENGER,V. "Unified control methods in dosimetry for high activity irradiation facilities in Hungary", High Dose Measurements in Industrial Radiation Processing, Technical Reports Series No. 205, IAEA, Vienna (1981).

[4] Dvornik I., The ethanolchlorobenzene dosimeter in manual on Radiation Dosimetry (Eds Holm N.W. and Berry R.J.) p. 345 Dekker, NewYork (1970).

[5] AVACO, C.M., ALMEİDA, C.J., ANDRADE, M.E., KOVACS, A., Dosimetry Commissioning for an Industrial Co^{60} Gamma Radiation Facility, Appl. Radiat. Isot. **42** No. 12 (1991) 1185–1188.

[6] DEMİREZEN Ü., "Irradiation Efficiency calculation of ındustrial gamma ırradiation plant.", VI. National Congress of Nuclear Science and Technology,Uludag University Bursa,Turkey (1993) (in Turkish).

METHODS TO VERIFY ABSORBED DOSE OF IRRADIATED CONTAINERS AND EVALUATION OF DOSIMETERS

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Abstract. The research on dose distribution in irradiated food containers and evaluation of several methods to verify absorbed dose were carried out. The minimum absorbed dose of treated five orange containers was in the top of the highest or in the bottom of lowest container. D_{max}/D_{min} in this study was 1.45 irradiated in a commercial ^{60}Co facility. The density of orange containers was about 0.391g/cm^3. The evaluation of dosimeters showed that the PMMA-YL and clear PMMA dosimeters have linear relationship with dose response, and the word NOT in STERIN-125 and STERIN-300 indicators were covered completely at the dosage of 125 and 300 Gy respectively.

INTRODUCTION

Agricultural exports including fresh and dry fruits and tree nuts provide important sources of foreign exchange for many developing countries. But the major importing countries such as Australia, Japan and USA have strict regulations for plant protection and quarantine. With the ban of ethylene dibromide in 1980s and limited use of methyl bromide in the near future, the two main fumigants used for controlling insect infestation including quarantine purposes, countries which depend on fumigation to overcome trade barriers in fresh fruits and other agricultural products will have to search urgently for alternative treatments to maintain their trade[1]. Irradiation, as one of the alternative treatment, is being increasingly recognized by authorities and organizations as an effective method for insect disinfestation of quarantine treatment[2]. The studies on quality control of irradiation especially methods to verify the absorbed minimum dose in irradiated food containers is needed[3,4].

MATERIALS AND METHODS

Materials: Orange containers were bought from market in November. PMMA-YL dosimeter, developed by Dr. Tang of the Institute for Application of Atomic Energy; clear PMMA sheets were supplied by the Pak Poly Industries (Pvt) Limited, Pakistan; STERIN indicators were provided by STERIN company, USA.

^{60}Co irradiation source in Irradiation Center of the Institute for Application of Atomic Energy was used for irradiation treatment. UVVIS Spectrophotometer WFZ800D3A was used for measuring the optical density of the dosimeters.

Dose distribution in irradiated orange containers: Dose distribution in irradiated orange containers was measured as follows in 1995: A total of 66 PMMA-YL dosimeters were placed in five orange containers together with standard Fricke dosimeter, 3 in a row. Nos 1–9, 19–24, 31–36, 43–48, 55–60 dosimeters were attached in one side of the container. Dosimeters Nos 10–18, 25–30, 37–42, 49–54, 61–66 were placed in the middle of the container. Dosimeters Nos 14, 26, 38, 50 and 62 were in the center of container 1, 2, 3, 4 and 5 (Figure 1). Five containers were placed played in a row, with container 1 on the top and container 5 on the bottom. The samples were irradiated static by ^{60}Co irradiator. The absorbed dose were planned to be 150 Gy.

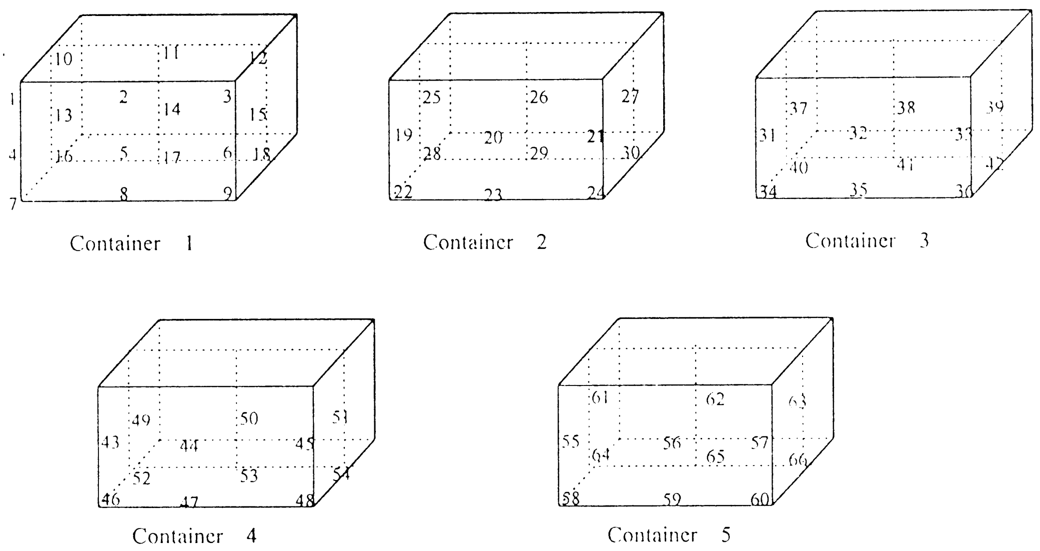

FIG. 1 Dosimeters position (166) in standardized orange containers, 1995.

Density of orange containers: oranges were packaged in paper cartons. The size of carton was 42 × 32 × 33 cm^3 .

The containers were turned 180° once during irradiation. The optical density of PMMA-YL dosimeter were measured at 530 nm before and after irradiation. The Fricke dosimeter was prepared and measured by standard method[5].

Density of orange containers: oranges were packaged by paper cartons. The size of carton was 42×32×33 cm^3.

Evaluation of dosimeters

1. Evaluation of PMMA dosimeter: PMMA-YL is a narrow strip of 10×40 mm^2 and 13 mm thickness. The color of dosimeter changes from yellow to red or dark red after irradiation, The wave length of 530 nm was used for the measurement. The dose range was 50–1000 Gy. The clear PMMA dosimeters include five sheets of 11×30 mm^2 pieces in 2, 4, 6, 8 and 10 mm thickness which are to be studied at dose level of 0.1–1.0 kGy.

Dose response of PMMA dosimeter: Six pieces of PMMA-YL dosimeters were irradiated at doses of 75, 100, 150, 200, 250 and 300 Gy in a ^{60}Co Gamma source. The thickness of dosimeters was 2.39–2.40 mm. Fricke dosimeter which was graduated by National Metrology Institute, was used as standard for calibration. The optical density of PMMA-YL dosimeter was measured at 530 nm before and after irradiation with spectrophotometer.

Six pieces of clear PMMA dosimeters were irradiated at doses of 100, 200, 300, 400 and 500 Gy in the ^{60}Co Gamma source. The thickness of dosimeters was 4.0 mm. The optical density of clear PMMA dosimeter was measured at 320 nm before and after irradiation with spectrophotometer.

PMMA dosimeters stored at different humidity: Out of ten pieces each of thickness 3.42–3.43 mm of PMMA-YL dosimeters and 4 mm of clear PMMA dosimeters, two each were kept in different containers filled with different proportions of glycerine and water to obtain the relative humidity of 60%, 71%, 80%, 91% and 94%. These dosimeters were irradiated at the dose rate of 2.0 Gy/min. The absorbed dose was 300 Gy for PMMA-YL and 500 Gy for clear PMMA. The bottles were stored in dark at room temperature. The optical density of PMMA-YL dosimeter were measured at 530 nm and clear PMMA dosimeter were measured at 320 nm before and after irradiation 0, 9, 43 and 81 days. For avoiding the effects of humidity, the irradiated dosimeters were also packaged in PE plastic bag.

2. Evaluation of STERIN indicators: Two type of indicators were first irradiated by ^{60}Co source at a dose rate of 36.2 Gy/min. The dose 75 Gy, 100 Gy and 150 Gy were used for STERIN-125 indicator evaluation and 150 Gy, 300 Gy and 400 Gy for STERIN-300 evaluation. Two samples were irradiated for each treatment.

The STERIN-300 indicators were irradiated by the new irradiator in our institute. The indicator was placed with ampoules (Fricke dosimeter), in or out of four containers in one carrier of overhead conveyor system.

RESULTS

1. Dose distribution in irradiated orange containers

The dose distribution in containers filled with orange was measured by PMMA-YL dosimeter and Fricke dosimeter[5]. The results of absorbed dose in 66 points of five containers are listed in Table 1. The results of two dosimeters were almost the same. The lowest absorbed dose were in position Nos 10,11 and 12, the top middle of container 1 and the bottom of container 5. The highest dose was in the sides of container 2, 3 and 4. D_{max}/D_{min} were 1.34 from the data of Fricke dosimeter and 1.53 from the data of PMMA-YL dosimeter.

According to the practical guide of dosimetry for food processing[6], for static irradiation, at least one dosimeter is needed for one batch of treated food product. In our experiment, if one dosimeter was placed at position 9 (Figure 1), the lowest absorbed dose in these five containers will be: Absorbed dose at position 9 × R, where, R is the lowest absorbed doses in containers/absorbed dose at position 9 = 157.6/188.8 = 0.835.

The weight of five orange containers were 17.1 kg 17.3 kg, 16.8 kg, 17.1 kg and 17.4 kg respectively. The volume of container was 42 × 32 × 33 cm^3. The density of orange containers was about 0.379 g/cm^3.

2. Evaluation of dosimeters

1. Evaluation of PMMA dosimeters: The dose response of PMMA-YL dosimeter is shown in Fig. 2. The coordinate curve shows the straight line relationship between the absorptive dosage and the optical density (OD) of the dosimeter. The relative coefficient R^2 = 0.9792.

The dose response of clear PMMA dosimeter is shown in Fig. 3. The curve shows the straight line relationship between the absorptive dosage and the optical density (OD) of the dosimeter. The relative coefficient R^2 = 0.9024.

TABLE 1. DOSE DISTRIBUTION IN ORANGES CONTAINERS MEASURED BY PMMA-YL AND FRICKE DOSIMETER

No.	Fricke	PMMA-YL	No.	Fricke	PMMA-YL	No.	Fricke	PMMA-YL
1	182.3	187[1]	23	199.5	172	45	202.6	214
2	178.1	166	24	202.3	169	46	207.1	207
3	179.2	161	25	179.2	150	47	200.0	212
4	178.9	161	26	175.0	149	48	209.1	213
5	179.2	164	27	174.8	151	49	199.5	189
6	178.7	167	28	195.8	191	50	186.0	173
7	186.0	160	29	193.9	177	51	183.2	171
8	182.3	164	30	195.3	181	52	183.8	178
9	188.8	167	31	205.4	210	53	184.0	170
10	157.0	145	32	200.3	192	54	184.6	190
11	157.6	144	33	209.6	208	55	185.7	200
12	157.9	140	34	210.1	211	56	203.4	192
13	167.7	147	35	207.6	192	57	189.1	193
14	161.0	143	36	208.2	192	58	181.5	185
15	162.7	146	37	187.7	167	59	176.4	170
16	179.0	151	38	187.7	160	60	188.2	173
17	178.4	152	39	180.9	161	61	164.9	174
18	179.8	156	40	197.2	196	62	163.5	176
19	197.8	179	41	199.9	199	63	168.3	180
20	197.2	175	42	199.8	201	64	163.2	153
21	197.8	171	43	201.7	201	65	163.8	151
22	202.6	180	44	201.2	189	66	163.8	148

Note: The data normalized by thickness.

The effects of relative humidity on the optical density of day 0, 9, 43 and 81 are shown in Fig. 4. The optical density changes of irradiated PMMA-YL dosimeters in different humidity was not much influenced by humidity in the experiment, but the optical density in 80% humidity was lower than others. After ten days storage, the optical density was stable at 60% and 71%, but lower at 80%, 91% and 94% humidity. This change was more obvious after 43 days storage and the optical density of 81 days samples was faded with the humidity. The optical density of plastic bag packaged samples showed almost no change, 0.3555 at day 0 and 0.3445 at day 81.

At days 0 and ten days later the optical density of irradiated clear PMMA dosimeters in different humidities showed no regular changes (Table 2).

2. Evaluation of STERIN indicators: The results for STERIN-125 and STERIN-300 indicators irradiated by four different doses were as follows. The STERIN-125 indicators changed to dark red after 75 Gy treatment and NOT word could be recognized clearly. At the dosage of 100 Gy the color of indicator got darker but the word NOT still could be seen. When the irradiation dose at/and more than 125 Gy, the color of indicator was black and the word NOT could not be seen at all. The results for STERIN-300 were similar with STERIN-125. The color of indicator also changed completely at 300 Gy and higher dose. The results were same after six months' storage.

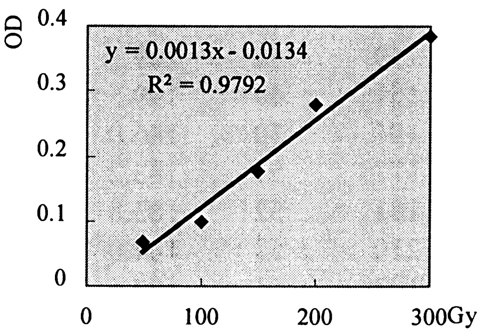

FIG. 2. Dose response of PMMA-YL dosimeters.

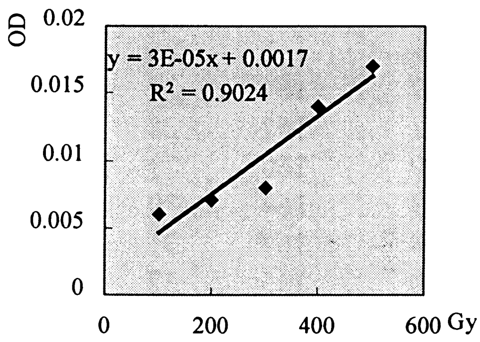

FIG. 3. Dose response of clear dosimeters.

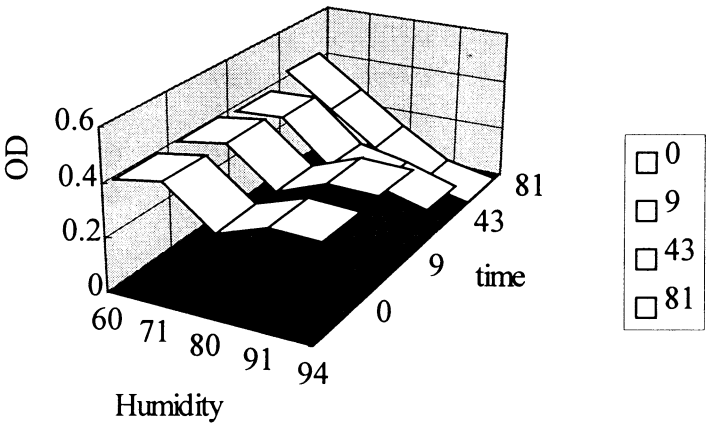

FIG. 4. The effects of different humidity on storage of irradiated PMMA-YL dosimeters.

TABLE 2. THE OPTICAL DENSITY CHANGES OF THE CLEAR PMMA DOSIMETERS AT DIFFERENT HUMIDITY AND AFTER 0, 10 DAYS STORAGE

humidity(%)	60	71	80	91	94
0 day	0.024	0.021	0.010	0.018	0.024
10 days storage	0.029	0.032	0.016	0.024	0.017

The results for STERIN-300 irradiated in and on four containers in one carrier of overhead conveyer system are shown in Table 3. When the absorbed dose measured by Fricke dosimeter was lower than 300 Gy, the NOT word in STERIN-300 indicator were shown clearly. When absorbed dose was higher than 300 Gy, the NOT word in STERIN-300 indicator changed to dark and masked the letter NOT. But the NOT word still could be seen in one position but the Fricke dosimeter there showed the absorbed dose of 332 Gy.

Conclusion

The minimum absorbed dose of treated five orange containers was in the top of the containers. D_{max}/D_{min} in this study was 1.451. The dose in a reference position (position 9) could be related to the minimum dose by a factor 0.835.

TABLE 3. IRRADIATION EFFECTS OF STERIN-300

STERIN-300	Fricke	STERIN-300	Fricke	STERIN-300	Fricke
1	247	++	398	++	377
	254	++	396	++	401
	248	++	391	+	332
++	384	++	378		264
++	417	++	394		272
++	376	++	392		262

Note: “” NOT word could be seen clearly.
“++” NOT word could not be seen.
“+” NOT word could be seen but not so clearly.

The evaluation results of two PMMA dosimeters show that the PMMA-YL dosimeter was useful at the 0–300 Gy irradiation. The absorbed dose was not much influenced by the relative humidity of lower than 70% after storage of 10 days. In order to avoid the effects of humidity, the PMMA-YL dosimeter with plastic package was suggested. For clear PMMA dosimeter, it seemed that 0–500 Gy absorbed dose was too low to be measured, the relative coefficient was only 0.9070. STERIN indicator appears to be adequate for a visual identification of 125 or 300 Gy irradiation.

ACKNOWLEDGEMENT

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REFERENCES

[1] Loaharanu, P., International trade in irradiated foods: regional status and outlook, Food Technology **43** (7) (1989) 77–80.
[2] FAO/IAEA, Co-ordinated Research Programmes, Food Irradiation Newsletter 8(2) (1986) 14–27.
[3] Thakur, B.R., Singh, R.O., Combination Processed in Food Irradiation, Trends in Food Science & Technology Vol. 6 (1995).
[4] Heather, N.W., Irradiation as a Quarantine Treatment of Agricultural Commodities Against Arthropod Pest IAEA-SM-327/58, IAEA, Vienna (1994) 627–639.
[5] Standard Method for Using the Ferrous Sulfate (Fricke) Dosimeter to Measure Absorbed Dose in Water, GB. 1989:13989.
[6] FAO/IAEA, Food Irradiation Newsletter 20(2) (1996) 14–19.

INVESTIGATING PHYSIOLOGICAL METHODS TO DETERMINE PREVIOUS EXPOSURE OF IMMATURE INSECTS TO IONIZING RADIATION

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Abstract. Effect of gamma radiation on phenoloxidase activity in codling moth, *Cydia pomonella* L., larvae was investigated. Phenoloxidase activity was determined spectrophotometrically by measuring the increase in optical density at 490 nm, or by observing the degree of melanization in larvae killed by freezing. Results showed that, in unirradiated larvae, phenoloxidase activity could be detected in 7 day old larvae and activity continued to increase throughout the larval stage. This increase was not observed when larvae were irradiated with a minimum dose of 50 Gy during the 1st week of their development. However, irradiating larvae in which enzyme activity was already high (24 week old) did not eliminate the activity but reduced further increase. Larval melanization studies were in general agreement with the results of the phenoloxidase assay.

1. INTRODUCTION

The codling moth, *Cydia pomonella* L., is a pest of pome and stone fruits throughout most deciduous fruit growing areas of the world. Its larvae infest apples, pears, walnuts, and many other deciduous fruit crops and causes hundreds of millions of dollars in losses to the fruits industry every year. This species is also of quarantine importance in Japan, Korea, Taiwan, Brazil, and parts of China [1] and strict quarantine measures are applied to prevent its entry and/or establishment in these countries. The insect overwinters as diapausing larvae in cocoons which, sometimes, are present within the shell of inshell walnuts. Therefore, inshell walnuts could not be exported to countries where codling moth is quarantined. The insect could also be transmitted as eggs or larvae on or in fruits. As a result, quarantine regulations are imposed on fruits exported to codling moth free countries.

Ethylene dibromide (EDB) was accepted to disinfest nuts and fruits exported to codling moth free countries [2]. Concern over its carcinogenic, mutagenic and environmental effects led the US government to phase out this chemical and several other countries have also adapted the same policy [3].

Ionizing radiation has been proposed as a possible alternative to chemical fumigation against commodities subject to quarantine regulations because of infestation by insects pests and a generic dose of 300 Gy was recommended for any pest other than fruit flies [4]. Studies on the codling moth has indicated that gamma radiation is an effective treatment against larvae in apples [5, 6]. However, quarantine doses do not necessarily cause immediate death and larvae may continue to develop to the pupal stage. Consequently, if a living larva is detected in a shipment, it is very important to determine if it has been irradiated [7] and whether the minimum required dose has been received.

Several studies have been done to distinguish irradiated larvae from those not irradiated. The size of the supraoesophageal ganglion in relation to the proventriculus was reduced when eggs and young larvae of several Tephritid fruit fly species were exposed to ionizing radiation [8, 9, 10]. Tsiropoulos [11] found that melezitose metabolism in adults of the olive fruit fly, *Dacus oleae* (Gmelin), was affected by irradiation. Irradiated flies were unable to hydrolyse this sugar into its basic components (2 glucose and 1 fructose). He suggested that a hydrolytic enzyme produced in gut cells had become inactive because of the irradiation treatment. The enzyme phenoloxidase plays an important role in the melanization of insect cuticle through the conversion of aromatic quinones and its products into melanin [12]. The enzyme is also responsible for the darkening (melanization) of insect tissue at injury sites or after death [13]. Nation et al. [14] studied the effects of gamma radiation on phenoloxidase and melanization in the larvae of the Caribbean fruit fly, *Anastrepha suspensa* (Loew). They found that 20 Gy applied to the late egg stage or young larvae drastically decreased the activity of this enzyme in 3rd instars and prevented melanization in larvae killed by freezing. Similar results on the Mediterranean fruit fly, *Ceratitis capitata*, were also reported by Mansour and Franz [15, 16].

The objectives of this investigation were to study the effects of gamma radiation on melanization and phenoloxidase activity in *C. pomonella* larvae killed by freezing and to investigate the possibility of using the level of phenoloxidase in irradiated larvae as an indication of irradiation dose.

2. MATERIALS AND METHODS

2.1. Insects. Larvae used in this research were obtained from a colony of *C. pomonella* that has been reared on an artificial media similar to that reported by Brinton et al. [17]. The larvae were reared in plastic boxes (18 × 15 × 6 cm) at 28±2 °C, 40 60 % RH and a photoperiod of 16:8 (L:D). Under these conditions, the larval stage requires approximately one month.

2.2. Irradiation. Larvae were irradiated in the larval rearing medium in plastic bags. Irradiation was done in a cobalt-60 source (Issledova Gamma Irradiotor, Techsnabexport Co. LTD, RUS). Radiation doses between 25 and 100 Gy with 25 Gy intervals were used and an untreated group was held as a control. The mean dose rate over the period of these tests was approximately 83.30 Gy/minute. After irradiation, larvae were returned to the rearing room and when they reached the desired age, they were killed by freezing.

2.3. Phenoloxidase assay. Larvae were homogenized individually in 150 µl of 0.1 M phosphate buffer solution (pH 6.5) in a 1.5 ml test tube. The homogenate was centrifuged for 5 min at 15,000 Xg, after which the supernatant, containing the enzyme, was kept on ice until tested to avoid possible autooxidation. The substrate was 5 mg/ml of L-dihydroxyphenylalanine in 0.1 M phosphate buffer solution. Fifty microliteres of the supernatant were added to 0.95 ml of the substrate solution, mixed with a vortex mixer for few seconds, and incubated at 25°C for 5 min. As a result of oxidizing L-Dopa to Dopachrome [18], a red color was produced that was quantified by measuring light absorption at 490 nm with a spectrophotometer [19]. Phenoloxidase activity was expressed as the increase in absorbance at 490 nm under the conditions described above.

2.4. Melanization test. Larvae exposed to 50 Gy dose were removed from the freezer and placed on a white background for observation. The color change (darkening) caused by the melanization reaction was compared with unirradiated controls.

2.5. Data analysis. Data from these experiments were subjected to analysis of variance. Means were separated by Duncan's [20] multiple range test.

3. RESULTS

3.1. Phenoloxidase Activity in *C. pomonella* larvae

Phenoloxidase activity in codling moth larvae was determined spectrophotometrically. Frozen larvae were used to increase the activity of the phenoloxidase system [18]. Results with unirradiated larvae (Table 1) showed that the activity of phenoloxidase was very low during the first week of larval development. Enzyme activity, however, continued to increase throughout the 4th week (end of larval stage). These results agree with those reported by Hackman and Goldberg [21] and Mnasour and Franz [16] for several species of insects.

TABLE 1. PHENOLOXIDASE ACTIVITY (OD UNITS $\times 10^3$) IN *C. POMONELLA* LARVAE*

Age of larvae (days) when assayed	Activity ± SD (OD units $\times 10^3$)
7	023.4 ± 05.9[a]
14	210.3 ± 61.8[b]
21	321.6 ± 73.1[c]
28	533.8 ± 91.8[d]

*Data represent the average of 10 measurements on individual larvae. Means followed by the same letter in each column are not significantly different (P>0.05).

3. 2. Effect of gamma radiation on phenoloxidase activity

When one week old larvae were irradiated and tested as 2, 3, and 4 week old larvae, phenoloxidase activity was significantly affected. At a dose of 50 Gy and higher, enzyme activity was very low (Table 2). Statistical analysis indicates that irradiation significantly reduced the level of phenoloxidase in examined larvae ($P < 0.001$). Comparisons between means showed that phenoloxidase activity was significantly reduced at a dose as low as 25 Gy ($P < 0.01$) and this reduction reached its maximum at 50 Gy dose.

TABLE 2. EFFECTS OF GAMMA RADIATION ON PHENOLOXIDASE ACTIVITY (OD UNITS $\times 10^3$±SD) IN *C. POMONELLA* LARVAE*

Dose	Assay days		
(Gy)	14	21	28
0	210.8 ± 62.3[a]	321.5 ± 73.3[a]	534.6 ± 90.8[a]
25	102.9 ± 23.6[b]	135.0 ± 33.4[b]	227.8 ± 45.9[b]
50	023.0 ± 09.0[c]	031.8 ± 08.3[c]	037.6 ± 07.6[c]
75	024.1 ± 05.0[c]	037.3 ± 05.9[c]	033.0 ± 09.6[c]
100	030.5 ± 10.7[c]	031.1 ± 07.0[c]	035.1 ± 10.0[c]

*Data represent the average of 10 measurements on individual larvae. Means followed by the same letter in each column are not significantly different (P >0.05).

3.3. Effect of age on phenoloxidase sensitivity to gamma radiation

Data on the effect of gamma radiation on phenoloxidase activity measured in 4 week old larvae, after irradiation as 1, 2, and 3 week old larvae showed that irradiation drastically reduced phenoloxidase activity, especially when larvae were treated during the 1st week (Table 3). However, when larvae were treated as 23 week old, the effect of radiation was less severe.

TABLE 3. EFFECTS OF GAMMA RADIATION ON PHENOLOXIDASE ACTIVITY (OD UNITS × 10^3±SD) IN MATURE *C. POMONELLA* LARVAE IRRADIATED AS 1, 2, AND 3 WEEK OLD AND EXAMINED AS 4 WEEK OLD*

Dose (Gy)	Irradiation days		
	7	14	21
0	500.8 ± 95.0[a]	500.8 ± 95.0[a]	500.8 ± 95.0[a]
25	121.2 ± 14.4[b]	201.6 ± 33.7[b]	270.0 ± 35.9[b]
50	025.3 ± 04.7[c]	121.5 ± 42.3[c]	204.3 ± 29.9[c]
75	019.6 ± 04.5[c]	154.2 ± 34.7[c]	217.4 ± 33.5[c]
100	022.0 ± 05.4[c]	132.5 ± 43.6[c]	211.9 ± 28.2[c]

*Data represent the average of 10 measurements on individual larvae. Means followed by the same letter in each column are not significantly different (P >0.05).

3.4. Effect of gamma radiation on melanization

Results of the larval melanization study (Table 4) agreed with results of the phenoloxidase assay. Untreated 1 week old larvae showed a clear lack of melanization, whereas 2, 3 and 4 week old larvae turned, partially or completely black after 2 hours of incubation at room temperature. In comparison with the unirradiated control, 24 week old larvae that had been irradiated as 1 week old larvae, particularly those that had been exposed to a dose of 50 Gy or higher, showed no melanization. However, when later stages were treated, irradiation did not prevent melanization in most of the examined specimens.

TABLE 4. RESULTS OF THE LARVAL MELANIZATION STUDIES*

Age of larvae (days) when irradiated	Age of larvae (days) when examined		
	14	21	28
7	+/	+/	+/
14		+/±	+/±
21			+/+

*Fifty larvae from each age group exposed to a dose of 50 Gy were examined.
+, melanized; _, not melanized; ±, inconclusive.

4. DISCUSSION

The irradiation dose of 300 Gy, as currently recommended for quarantine treatment against all insects other than fruit flies of the family Tephritidae [4], does not prevent *C. pomonella* eggs or larvae from developing to the pupal stage [5, 6]. Consequently, a method is needed to establish with certainty that larvae detected in imported fruits were indeed irradiated and exposed to the minimum required dose that prevents their development into sexually reproductive adults. In a search for a test system, Nation et al. [14] and Mansour and Franz, [15, 16] suggested that the effect of radiation on enzymes, particularly phenoloxidase, present in insect larvae, be examined. In this report, we present results of research conducted on the effects of gamma radiation on phenoloxidase activity and melanization in codling moth larvae killed by freezing.

Examination of phenoloxidase activity in codling moth larvae showed that the activity of this enzyme can be detected in unirradiated larvae starting with the 1st week and increases until it reaches its maximum immediately before pupation. These results agree with those reported by Hackman and Goldberg [21] and Mnasour and Franz [16] for other insect species. Irradiation during the first week, with a minimum dose of 50 Gy, reduced this enzyme activity to levels close to the detection level and prevented melanization. However, when 23 week old larvae were irradiated, the reduction in both phenoloxidase activity and melanization was not sufficiently pronounced to allow visual discrimination between irradiated and unirradiated larvae. These results indicate that measuring the activity of this enzyme in *Cydia pomonella* larvae can be used as an indication of irradiation treatment provided that the limits mentioned above are observed. Failure of codling moth larvae to melanize if exposed to a minimum dose of 50 Gy during the 1st week can also be used as an indicator of irradiation treatment. However, since a dose of 50 Gy affects the formation of phenoloxidase in developing larvae and the required dose for quarantine treatment is 300 Gy (4) this method will not be suitable to verify that larvae received the minimum required dose.

REFERENCES

[1] Shel'deshova. G.G. 1967. Ecological factors determining distribution of codling moth, Laspeyresia pomonella L. (Lepidoptera: Tortricidae) in northern and southern hemispheres. Entomol. Rev. 46: 349–361.

[2] Anonymous. 1978. Ministry of Agriculture and Forestry (Japan). Notification No. 38. Japan plant protection law enforcement regulations (January 10, 1978) 27–33.

[3] Anonymous. 1984. Revocation of tolerances for ethylene dibromide. Fed. Reg. 49: 220825.

[4] Anonymous. 1986a. Task force meeting on irradiation as a quarantine treatment, Chiang Mai, Thailand, 1986. Food Irradiation Newsletter 10: 510.

[5] Burditt, A.K., and H.R. Moffitt. 1985. Irradiation as a quarantine treatment for fruit subject to infestation by codling moth larvae, pp. 87–97. In: J.H. Moy (Ed.), Proceedings, International Conference on Radiation Disinfestation of Food and Agricultural Products. University of Hawaii, Honolulu.

[6] Burditt, A.K., H.R. Moffitt and F.P. Hungate. 1985. Effects of gamma radiation as a quarantine treatment on the development of the codling moth larvae, pp. 38. In: International Symposium on Food Irradiation Processing. IAEA. Vienna.

[7] Anonymous. 1986b. Report of the FAO/IAEA research coordination meeting on the use of irradiation as a quarantine treatment of food and agricultural commodities, Chiang Mai, Thailand, 1986. Food Irradiation Newsletter 10: 1728.
[8] Jessup, A.J., C.G. Rigney, A. Miller, R.F. Sloggett, and N.M. Quinn. 1992. Gamma irradiation as a commodity treatment against the Queensland fruit fly in fresh fruit, pp. 1342. In: Use of irradiation as a quarantine treatment of food and agricultural commodities, Proceedings, Final Research Coordination Meeting, 1990, Kuala Lumpur, Malaysia. IAEA, Vienna.
[9] Rahman, R., C. Rigney and E. Busch Petersen. 1990. Irradiation as a quarantine treatment against Ceratitis capitata (Diptera: Tephritidae): anatomical and cytogenetic changes in mature larvae after gamma irradiation. J. Econ. Entomol. 83: 1449–1454.
[10] Rahman, R., A.D. Bhuiya, S.M.S. Huda, R.M. Shahjahan, G. Nahar, and M.A. Wadud. 1992. Anatomical changes in the mature larvae of two Dacus spp. following irradiation, pp. 133–139. In: use of irradiation as a quarantine treatment of food and agricultural commodities, proc. Final Research Coordination Meeting. 1990, Kuala Lumpur, IAEA, Vienna.
[11] Tsiropoulos, C.J. 1980. Carbohydrate utilisation by normal and gammasterilised Dacus oleae. J. Insect Physiol. 26: 633–637.
[12] Anderson, S.O. 1985. Sclerotization and tanning of the cuticle, pp. 59–74. In G. Kerkut and L. Gilbert [eds.], Comprehensive insect physiology, biochemistry and pharmacology, Vol. 3. Pergamon, Oxford.
[13] Salt, G. 1970. The cellular defense reactions of insects. Cambridge Monograph in Experimental Biology, No. 16. Cambridge University Press, London.
[14] Nation J.L., B.J. Smittle and K. Milne. (1995). Radiation induced changes in melanization and phenoloxidase in Caribbean fruit fly larvae (Diptera: Tephritidae) as the basis for a simple test of irradiation. Ann. Entomol. Soc. Am. 88: 201–205.
[15] Mansour M. and G. Franz. (1995). A rapid test for distinguishing irradiated from unirradiated medfly, Ceratitis capitata (Diptera: Muscidae), larvae, pp. 505–510. In: Fruit Flies of Economic Importance, 510 June 1994, St. Lucie press, FL, USA.
[16] Mansour M. and G. Franz. (1996). Effects of gamma radiation on phenoloxidase activity in Mediterranean fruit fly (Diptera: Tephritidae) larvae. J. Econ. Entomol. 89: (in press).
[17] Brinton, F. E., M.D. Proverbs and B.E. Carty. 1969. Artificial diet for mass production of the codling moth, Carpocapsa pomonella (Lepidoptera: Olethreutidae). Can. Entomol. 101: 577–584.
[18] Horowitz, N.H., and S.C. Shen. 1952. Neurospora tyrosinase. J. Biol. Chem. 197: 513–520.
[19] Bidochka, M.J., J.P. Gillespie, and G.G. Khachatourians. 1989. Phenoloxidase activity of acridid grasshoppers from the subfamilies Melanoplinae and Oedipodinae. Comp. Biochem. Physiol. 94B: 117–124.
[20] Duncan, D.B. 1955. Multiple range and multiple F tests. Biometrics 11: 124.
[21] Hackman, R.H., and M. Goldberg. 1967. The Odiphenoloxidase of fly larvae. J. Insect Physiol. 13: 531–544.

DEVELOPMENT OF FLUORESCENT, OSCILLOMETRIC AND PHOTOMETRIC METHODS TO DETERMINE ABSORBED DOSE IN IRRADIATED FRUITS AND NUTS

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Abstract. To ensure suitable quality control at food irradiation technologies and for quarantine authorities simple routine dosimetry methods are needed for absorbed dose control. Taking into account the requirements at quarantine locations these methods would require nondestructive analysis for repeated measurements. Different dosimetry systems with different analytical evaluation methods have been tested and/or developed for absorbed dose measurements in the dose range of 0.1–10 kGy. In order to use the well accepted ethanolmonochlorobenzene dosimeter solution and the recently developed aqueous alanine solution in small volume sealed vials a new portable, digital, and programmable oscillometric reader was developed. To make use of the availability of the very sensitive fluorimetric evaluation method, liquid and solid inorganic and organic dosimetry systems were developed for dose control using a new routine, portable, and computer controlled fluorimeter. Absorption or transmission photometric methods were also applied for dose measurements of solid or liquid phase dosimeter systems containing radiochromic dye agents, which change colour upon irradiation.

1. INTRODUCTION

The worldwide trade of agricultural products has shown continuous growth in the past decades and this tendency is increasing. Consequently the worldwide hygienic control of imported foods is of basic significance and requires high quality standardized control methods. Fresh and dried fruits, nuts, herbs and grains are often infested with insects, thus quarantine treatment is necessary for disinfestation. As the importance of fumigation as the most often applied method for insect control decreases significantly, the application of ionizing radiation has got new prospects recently. The correct use of radiation processing technologies requires correct dosimetry control methods, e.g. for dose distribution determination within the product packages controlling both the required minimum dose and the acceptable maximum dose. Quarantine authorities, on the other hand, often need simple routine methods to determine dose absorbed by the product on spot. The use of semiquantitative label dose indicators or simple, quick, easy to evaluate routine dosimeters with nondestructive analysis have got importance recently with respect to the increasing need of satisfying the regulatory inspectors at the importing stations to determine whether or not the irradiation for quarantine treatment was carried out correctly.

Fluorimetric, oscillometric and certain photometric methods possess the required simple and quick routine analysis applicable for dose determination.

2. EXPERIMENTAL

2.1. Chemicals and dosimeters

In order to determine absorbed dose applied during the gamma irradiation of the oscillometric, fluorimetric and photometric samples, ethanolmonochlorobenzene (ECB) dosimeter solution was prepared and used as described elsewhere (1). The calibration procedure was carried out using the regularly applied 2 cm^3 nominal volume glass ampoules and traceability to national standards was assured. In the course of the present study a new 1 cm^3 nominal volume vial was also introduced and checked for routine use in the same, i.e. 0.4–100 kGy dose range.

A new polymeric thin wafer (0.5 mm thickness), containing a microcrystalline dispersion of a proprietary optically stimulated fluor in a plastic matrix has also been developed by Sunna Systems Corporation (2) and studied also by us for routine dosimetry using a simple, tabletop fluorimeter developed in Hungary.

βnaphthylamine, quinoline and naphthalene2carboxylic acid were of reagents grade and were used as received.

Tetrazolium violet, blue tetrazolium chloride, nitroblue tetrazolium chloride, tetrazolium red and phosphomolibdic acid from Fluka were also of reagent grade and used without further purification.

Solid state scintillators containing organic fluors, such as 1phenyl3mesityl2pyrazoline, 3hydroxyflavone and 2(2hydroxyphenylbenzathiazole) were produced by Bicron Co. (USA).

2.2. Irradiation facilities

The liquid and solid dosimetry samples, studied with oscillometry, fluorimetry and photometry, were irradiated with different 60Co gamma irradiation facilities. Irradiations were carried out within the source cage in a calibrated position of the SLL01 type gamma source (3 PBq) of the Institute of Isotopes Co. Ltd. and with the Gammacell 220 type gamma irradiation facility (0.7 PBq) of the National Institute of Standards and Technology (Gaithersburg, USA).

The electron irradiations were carried out with the 4 MeV linear electron accelerator (LPR4 type, Tesla Vuvet, Praha, Czech Republic) of the Institute of Isotope and Surface Chemistry (Budapest, Hungary) using 2.6 μsec pulses, 25 cm scan width and 1 Hz scanning frequency.

2.3. Instrumentation

Oscillometric measurements were performed with the OK302/2 type oscillotitrator of Radelkis (Budapest, Hungary). The new 1 cm^3 volume ECB containing vials were tested with a new, digital oscillometric reader built recently by Sensolab Ltd. (Budapest, Hungary).

The fluorimetric investigations were carried out with the LS5 Luminescence Spectrometer (PerkinElmer), with the Clinifluor 88 PT routine fluorimeter (Institute of Isotopes Co. LTD) modified for our purposes and with the routine, programmable fluorimeter produced by Sensolab Ltd.

The spectrophotometric measurements were performed with the Jasco V550 UVVIS and the Cary 14 UVVIS spectrophotometers.

3. RESULTS AND DISCUSSION

3.1. Oscillometric investigations

Oscillometry is an electroanalytical method of conductivity measurements, where high frequency alternating current is applied to measure or follow changes in the composition of chemical systems. The ampoule containing the solution under test is placed either between the plates of a capacitor (capacitive cell) or inside the inductance coil (inductive cell) of an oscillator. The main advantage of the method is that the electrodes are not in direct contact with the solution, thus the analysis can be carried out in sealed ampoules too. Thus the method is nondestructive, making possible the quick, repeatable, routine dose evaluation of the irradiated dosimeters.

The ethanolmonochlorobenzene dosimeter solution is a well known reference and routine system for dose control in radiation processing in the 0.4–100 kGy dose range using oscillometric evaluation method (3). The original dosimeter system applies an analogue oscillotitrator for the evaluation of the irradiated solution. The nondestructive analysis makes possible the repeated evaluation of the irradiated dosimeters at any time after irradiation and according to our experiences irradiated ECB dosimeters can be evaluated in case of suitable storage 15 years after irradiation with a reproducibility of ±4%.

In order to carry out quick routine measurements for the present purpose i.e. at quarantine locations a new, portable, digital oscillometric reader was designed and built, suitable for measurement with both 2 cm^3 and 1 cm^3 nominal volume ampoules. Due to the necessity of using a small size dosimeter a new, 1cm^3 volume, sealed glass vial (diameter: 7.2 mm; wall thickness: 0.55 mm, produced by Chromacol, USA) was introduced to store the dosimeter solution. The new oscillometric reader, which can be connected to a PC, contains a builtin software making possible the calculation of absorbed dose by using the mathematical function of the calibration curve measured with the reader using ECB dosimeters calibrated previously. Since it was found earlier that the best fit was given by applying 3rd order polynomial this function is stored in the memory of the new reader.

The new system, i.e. the new reader and the 1 cm^3 vial, was tested with respect to routine application in the required dose range. The sealed vials were checked concerning their storage capabilities, i.e. the amount of liquid sealed in the vial was measured from time to time in order to test any leakage, but no change was observed after even 3 months storage. The diameter of the vial was also controlled and was found to vary between 6.9 mm and 7.4 mm. Thus, due to the diameter dependence of the oscillometric evaluation the vials must be selected and corrections have to be applied with respect to the calibration vials (7.1 mm ± 0.1 mm) similarly to the 2 cm^3 ampoules.

The vials were then irradiated in the 0.4–100 kGy dose range and measured with the new reader. The response of the system, i.e. the calibration curve is shown in Fig. 1. The results indicate, that the ECB solution can be used for dose evaluation with the new type oscillometric reader and applying the 1 cm^3 nominal volume vials in the same dose range as used with the previous analogue oscillometric system.

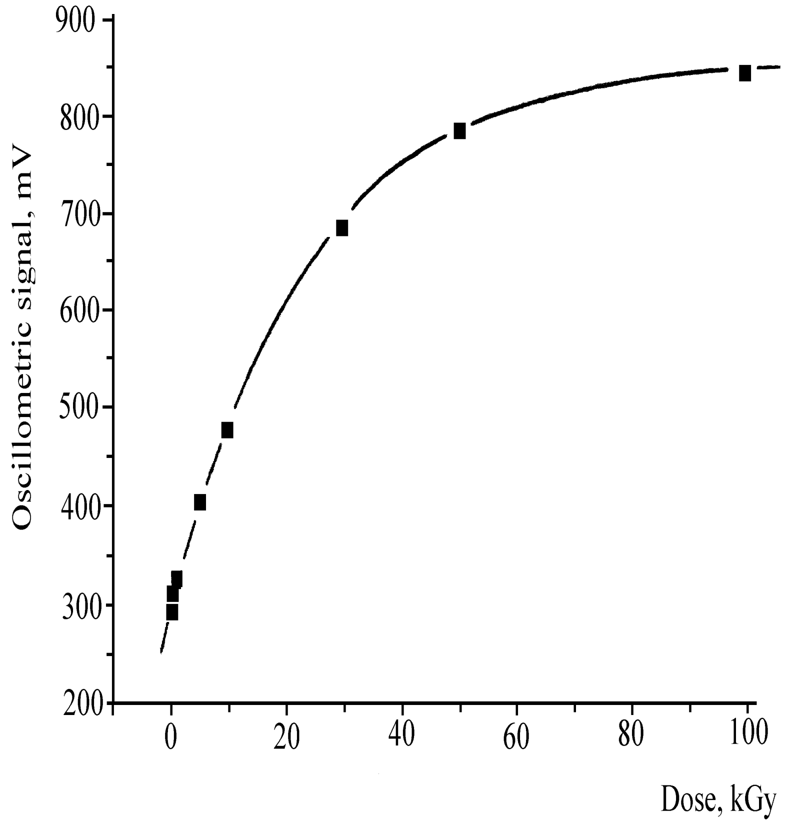

FIG. 1. Dose response of the oscillometric signal of ECB solution irradiated in 1 cm³ vials.

3.2. Fluorimetric investigations

Fluorimetry is a versatile method of dosimetry with the possibility of carriyng out measurements in a wide dose range. It is based on the measurement of OSL (optically stimulated luminescence), when a molecule — excited by UV or visible light — emits part of its energy in the form of light while returning to ground state. The intensity of the fluorescent light is related to the concentration of the fluorescent compound produced by radiation. In this case the original compound (i.e. the sample studied for possible use for dosimetry purposes) does not give fluorescence before irradiation, since the fluorescent compound is a radiation product [4].

It is also possible to study such a molecule, which originally contains a fluorescent compound (e.g. fluorescein or its derivative). This compound is then destroyed by radiation and the original intensity of the fluorescent light decreases. This decrease can again be the measure of absorbed dose.

In our investigations we have studied the possible use of some organic compounds like naphthalene1 and 2 carboxylic acids, βnaphthylamine, quinoline in aqueous solution as well as in solid matrix. By exciting the previously gamma irradiated basic aqueous solution of 20 mmol dm^3 naphthalene2carboxylic acid with 365 nm light a dose dependent emission was observed at 420 nm in the dose range of 0.1–100 kGy. In the case of gamma irradiated aqueous solutions of βnaphthylamine and quinoline precipitation was observed due to irradiation which became more and more significant with increasing dose. These compounds were then used in gelatin and polyvinylalcohol (PVA) and irradiated in the dose range of 0.1–50.0 kGy. The investigations concerning their applicability needs continuation.

Taking into account the simpler use of solid samples the possible use of originally fluorescent solid state plastic samples for dosimetry purposes was also investigated. Three different compounds, i.e. 1phenyl3mesityl2pyrazoline (PMP), 3hydroxyflavone (3HF) and 2 (2hydroxyphenyl) benzathiazole (HBT) in plastic matrix were irradiated in the dose range of 0.1–100 kGy and the excitation and emission spectra of the samples were measured. In the case of the PMP (excitation wavelength: 330 nm; emission wavelength: 425 nm) and 3HF (excitation wavelength: 355 nm; emission wavelength: 530 nm) (Fig. 2) the intensity of fluorescent light decreased with increasing dose, while in the case of HBT (excitation wavelength: 350 nm; emission wavelength: 517 nm) no significant change with respect to the fluorescent light intensity was observed. A simple routine fluorimeter (Clinifluor 88 PT produced by the Institute of Isotopes Ltd. Co., Budapest, Hungary) was found to be suitable for routine dose determination in the dose range studied.

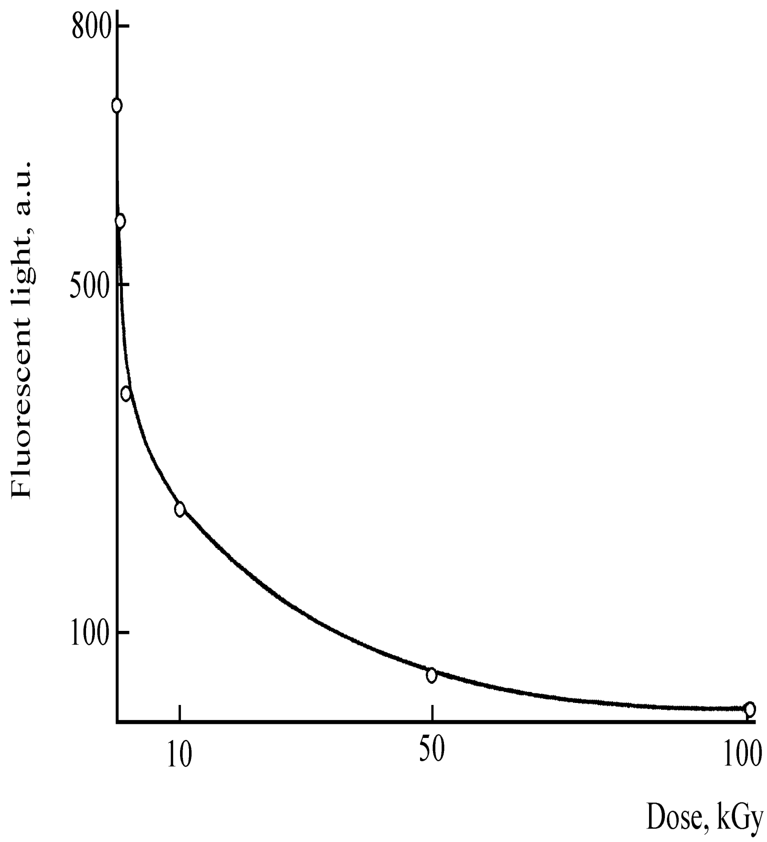

FIG. 2. Dose dependence of the fluorescent light originating from the BCF9950 fluor sample containing 3 hydroxyflavone.

Commercially available perspex (polymethylmetacrylate, PMMA) samples were irradiated in the dose range of 1–100 kGy and the fluorescent light intensity was measured at 480 nm (excitation wavelength: 350 nm). Since increasing fluorescent light intensity was observed with increasing dose it is assumed that a photofluorescent compound was produced upon irradiation. Due to the fact that various perspex samples are available commercially of different origin detailed investigations are needed with respect to stability and reproducibility when such samples are considered for dose measurements using fluorimetric method.

Optically stimulated luminescence dosimetry using inorganic microcrystalline solidstate fluors dispersed in a polymer matrix is a well known method for measuring low and high doses [2]. The potential applicability of a new film, the SUNNA dosimeter film, which contains an optically stimulated fluor in a plastic matrix, was also studied. The 0.25 mm thick film was irradiated in the 0.01–100 kGy dose range and the OSL signal was measured with a tabletop, routine fluorimeter exciting the irradiated 2.0 cm × 3.8 cm film samples at 450 nm. The emitted light was measured at 650 nm and 700 nm respectively. The response of the gammaray irradiated films was measured 1 h and 24 h after irradiation. There is a linear response up to about 40 kGy (Fig. 3), while at higher doses (30–100 kGy) significant sublinearity appears. This sublinearity, however, becomes nearly linear after 24 h storage. According to our investigations this OSL signal is not changed by many readout cycles and it is also not affected by light and humidity. The film has got a wide dynamic range, it is dose rate independent, is simple to use and is rugged. On the other hand it shows a slight temperature dependence and especially at high doses some instability for the first few hours after irradiation [2].

For routine measurements a programmable digital fluorimeter has been developed, which reads and evaluates the SUNNA film taking into account the calibration curve stored in its memory, but connecting it to a PC is also available.

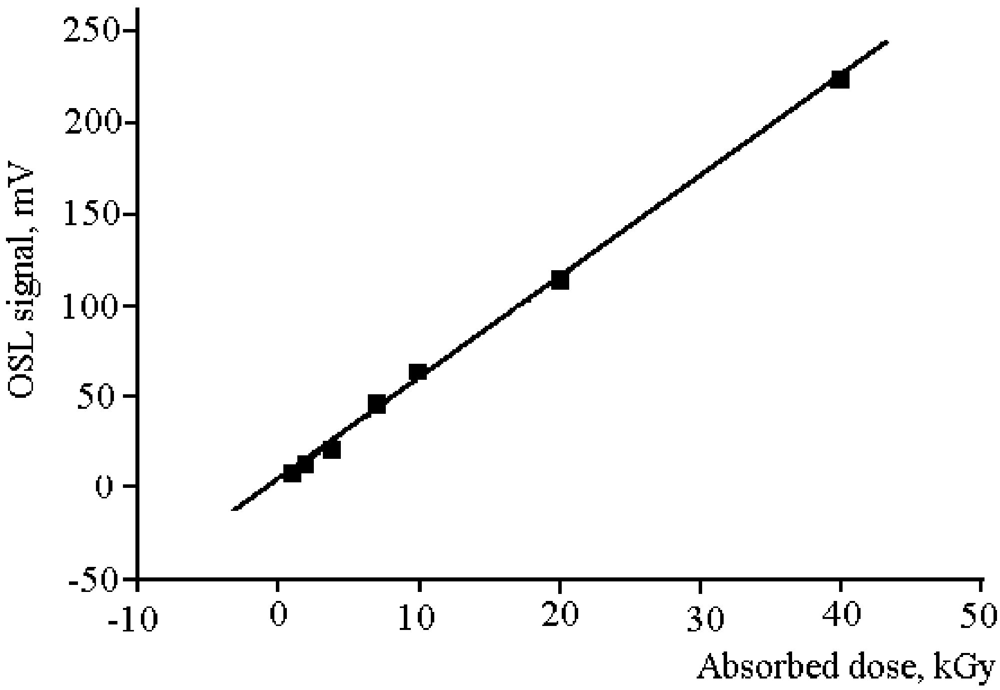

FIG. 3. Photofluorescent gammaray response of the 0.25 mm thick SUNNA dosimeter film measured one hour after irradiation at 650 nm.

3.3. Photometric investigations

Various types of radiochromic dye films are in regular use for absorbed dose measurements in radiation processing. The chemical background of these systems is the color change induced by radiation, i.e. the change from colorless to strongly colored takes place during irradiation and there is a relationship between this color change and the dose absorbed in the system. These radiochromic dye systems can be applied for dosimetry purposes both in liquid and solid state (films) [5].

Taking into account the request for developing a "label" system for dosimetry purposes different type of tetrazolium salts have been studied for potential application.

3.3.1. Nitroblue tetrazolium

Detailed investigations have been carried out to study the radiation chemical characteristics of this dye in order to determine the most suitable composition of the dye solution to be used in a solid host material. It was found that upon irradiation — due to a reductive process — highly coloured mono and/or diformazan radiolysis products form showing a dose and pH dependent absorption in the 500–600 nm region (Fig. 4). The formation of this product is much more pronounced at basic pH, i.e. at above pH ~10 and when the aqueous solution contains also approx. 0.1–0.5 mol dm^3 ethanol. In air saturated, aqueous solutions at basic pH together with the lilac colour of the irradiated solution precipitation takes place, while this precipitate gets dissolved in those solutions which contain e.g. ethanol.

Analyzing the results of the dose effect study our conclusion was, that aqueous solutions containing 1 mmol dm^3 dye can be used for dose measurements in the range of 0.01–1.0 kGy, where linear relationship between absorbance of the formazan formed (670 nm) and absorbed dose was found (Fig. 5).

3.3.2. Other tetrazolium salts

The potential use of aqueous solutions of blue tetrazolium chloride and tetrazolium violet was also studied for dosimetry application. The solutions containing 10 mmol dm^3 solute were irradiated in the dose range of 0.01–5.0 kGy and similar absorption spectra to the one observed in the case of nitroblue tetrazolium chloride were found indicating the same type of radiolysis product formation. These solutions are now under investigation concerning their potential use for dose determination in different concentrations, composition and pH.

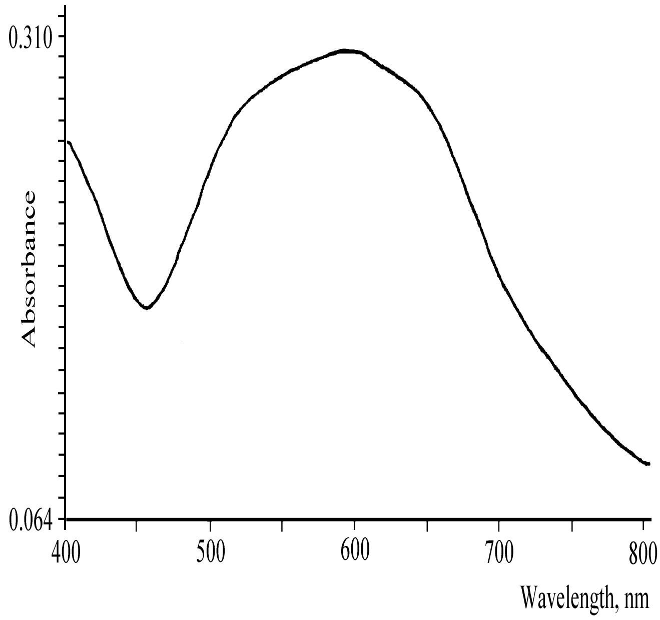

FIG. 4. Absorption spectrum of the irradiated nitroblue tetrazolium solution containing 0.5 mol dm^3. tertbutanol (pH = 10.3).

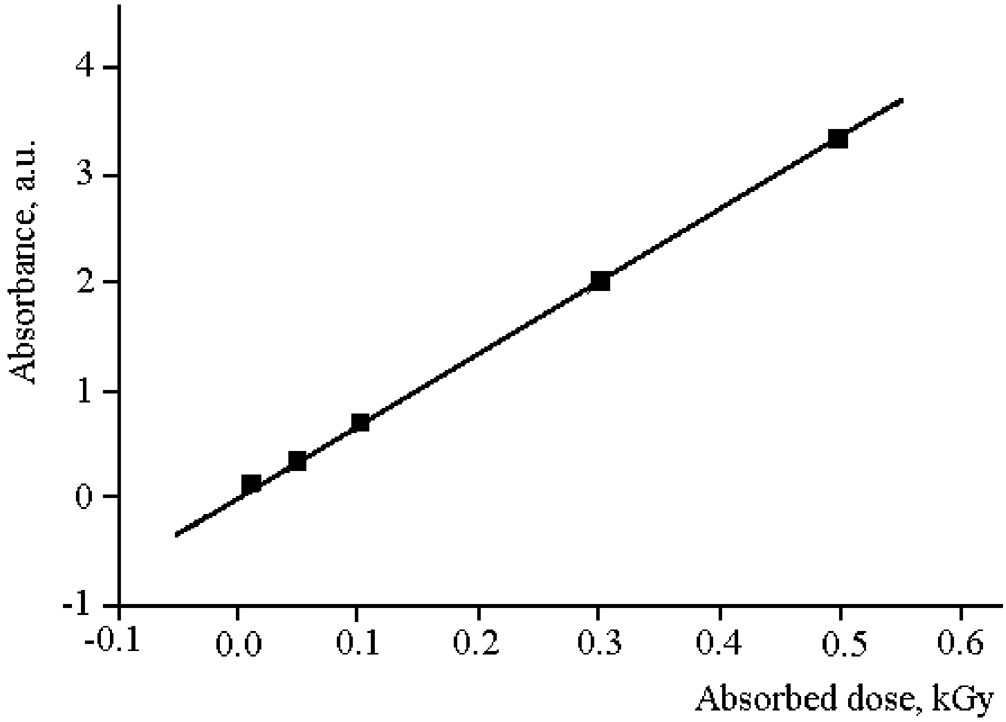

FIG. 5. Dose response of the absorbance (670 nm) of the irradiated aqueous nitroblue tetrazolium solution (pH = 11). (The overall uncertainty of the measurement at one standard deviation is estimated to be ±6%, as based on the statistical evaluations, i.e. type A uncertainties.)

3.3.3. Solid formulation of dye compounds

In order to develope solid phase label type systems both the fluorimetric and the photometric solutions studied so far were investigated with respect to their formulation in solid host materials. In the course of the first measurements gelatin and polyvinilalcohol (PVA) were used.

Aqueous solutions of quinoline and βnaphthilamin were mixed with gelatin and polyvinilalcohol (PVA) solutions respectively and the resulting dried samples were irradiated in the dose range of 0.1–50 kGy. The color gels formed upon irradiation are under detailed investigation for potential dose evaluation using both photometric and fluorimetric analysis.

Aqueous tetrazolium red solutions were also mixed with polyvinilalcohol solutions, dried and irradiated in the dose range of 0.1–10 kGy. The red colored gels formed upon irradiation are also studied for potential use as dose indicators and/or label dosimeters.

4. CONCLUSIONS

The oscillometric analysis of the ECB solution filled in the 1 cm^3 vials and analyzed with the new oscillometric reader gives more convenient application concerning nondestructive readout possibilities even in the low dose range from 0.4 kGy.

The application of the new SUNNA dosimeter film with OSL analysis provides convenient evaluation of as low as 10 Gy doses with repeated readout possibilities similarly to the oscillometric analysis. It is important to note that due to its size it offers good dose evaluation possibilities for electron beam applications too.

Tetrazolium salts both in aqueous solution and in gel and film form show promise for dose determination in the dose range of 0.01–10 kGy using both photometric and in certain cases fluorimetric evaluation method.

REFERENCES

[1] Horvath, Zs., Banyai, E., Foldiak, G., The Oscillometric EthanolChlorobenzene Dosimeter, Radiochim. Acta 13 (3) (1970) 150.

[2] McLaughlin, W.L., Miller, S.D., Saylor, M.C., Kovacs, A., Wojnarovits, L., An Inexpensive Mass Produced High Dose Polymeric Dosimeter Based on Optically Stimulated Luminescence, Radiat. Phys. Chem. (in press).

[3] Standard Practice for Use of the EthanolChlorobenzene Dosimeter System, ASTM Standard E 1538 93.

[4] Berlman, I.B., Handbook of Fluorescence Spectra of Aromatic Molecules, 2nd Ed., Academic Press, New York (1971).

[5] McLaughlin, W.L., Chen, Y.D., Soares, C.G., Miller, A., VanDyke, G., Lewis, D.F., Nucl. Instr. Methods in Phys. Res. (1995), A302, 165.

COMMERCIAL POWER SILICON DEVICES AS POSSIBLE ROUTINE DOSIMETERS FOR RADIATION PROCESSING

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Abstract. The use of silicon devices as possible radiation dosimeters has been investigated in this study. A bipolar power transistor in TO126 plastic packaging has been selected. Irradiations, with doses in the range from 50 Gy up to 5 kGy, have been performed at room temperature using different radiation sources (^{60}Co g source, 2.5, 4 and 12 MeV electron accelerators). Few irradiations with g rays were also done at different temperatures. A physical parameter, T, related to the charge carrier lifetime, has been found to change as a function of irradiation dose. This change is radiation energy dependent. Long term stability of the electron irradiated transistors has been checked by means of a reliability test ("high temperature reverse bias", HTRB) at 150 °C for 1000 h. Deep level transient spectroscopy (DLTS) measurements have been performed on the irradiated devices to identify the recombination centres introduced by the radiation treatment. The results obtained confirm that these transistors could be used as routine radiation dosimeters in a certain dose range. More work needs to be done particularly with g rays in the low dose region (50–200 Gy) and with low energy electrons.

1. INTRODUCTION

The use of silicon devices as possible radiation monitors has been considered since many years because of the effect of ionizing radiation on the physical and electrical properties of these devices [1]. These effects can be summarized as follows:

(1) production of a transient electric current across the pn junction during irradiation due to diffusion of electronhole pairs in the electric field of the depletion layer of the semiconductor; a permanent damage to the silicon crystal structure with formation of point defects, which creates generationrecombination centres that affect the charge carrier lifetime and all the related electrical parameters.

While in the first case the device can be used for real time doserate measurements [2], the permanent damages in the crystal structure permit measurement of dose in a wide range in terms of radiationinduced changes in electrical characteristics [3, 4]. In this latter case such devices have been proposed for dose measurements and used in free flowing product being transported in bulk through a radiation field [5, 6]. Looking into our work done in the past years on the effects of gamma and electron irradiation on silicon power devices [7] and on the basis of the experience acquired in this field it was decided to investigate the use of some commercial power transistors, available on the Italian market, for radiation dosimetry.

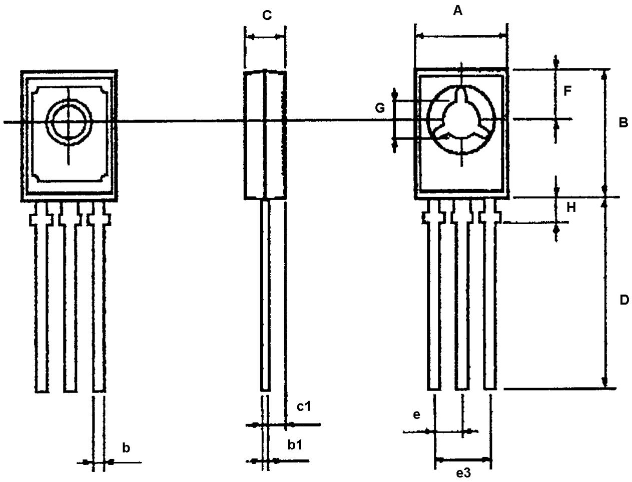
2. EXPERIMENTAL METHODS

2.1. Semiconductor device

A commercial bipolar transistor type BULT118 in TO126 or SOT32 plastic packaging, manufactured by SGSThomson, was selected. It is a high voltage fastswitching npn power transistor which is used in electronic ballasts for fluorescent lighting and in general for medium switching power applications. It is fabricated by using a multiepitaxial mesa technology from a 70 Ωcm CZ silicon substrate. The sensitive area is equal to 2.3 mm^2. Its mechanical data are reported in Table I.

TABLE I. SOT32 MECHANICAL DATA

DIM.	mm			DIM.	mm		
	MIN.	TYP.	MAX.		MIN.	TYP.	MAX.
A	7.4		7.8	D		15.7	
B	10.5		10.8	e		2.2	
b	0.7		0.9	e3		4.4	
b1	0.49		0.75	F		3.8	
C	2.4		2.7	G	3		3.2
c1		1.2		H			2.54



2.2. Irradiation and irradiation sources

All the devices have been irradiated at room temperature using different radiation sources located in different places. girradiations have been performed with the transistors enclosed in a plastic chamber with a wall thickness of 0.4 g/cm^2 which is adequate to establish electron equilibrium for ^{60}Co g rays. The irradiation facilities used were the ^{60}Co Nordion Gammacells 220 of the FRAE Institute in Bologna and of the IAEA laboratory in Seibersdorf, having dose rates of 50 Gy/min and 2.5 and 53 Gy/min respectively. Two other series of g irradiations have been done at the Institute of Isotopes in Budapest and at NIST, Washington using the local ^{60}Co g irradiators having dose rates of 33 Gy/min and 172.7 Gy/min respectively. The irradiation at NIST were performed at three different temperatures: 0°C, 25°C and 50°C. The temperature in the other g facilities was between 30 and 35°C.

Electron irradiations have been carried out in Bologna, in Budapest and in Strasbourg using pulses of electrons from an Lband 12 MeV, a Tesla Model LPR4 MeV linear accelerators and a 2.5 MeV Van de Graaff accelerator respectively. The machine parameters used for these irradiations are reported in Table II.

TABLE II ACCELERATOR CHARACTERISTICS

Equipments Characteristics	Tesla LPR4 Linac (Magnetron) Institute of Isotopes Budapest, Hungary	LBand Vickers Linac (Klystron) CNR – FRAE Bologna, Italy	Van de Graaff AERIAL Strasbourg, France
Max Beam Energy	4 MeV	12 MeV	2.5 MeV
Av. Beam Energy	3.8 MeV	8.2 MeV	2.2 MeV
Av. Current	20 mA	85 mA	50 mA
Pulse Duration	2.6 ms	2 ms	Continuous
Repetition Rate	50 p.p.s.	50 p.p.s.	Scan rate 20 Hz

From four to six transistors for each dose, in groups of two, three, four or six, have been irradiated at the same time.

2.3. Dosimetry

The dose rate of the Gammacell in Bologna has been measured in a fixed position (i.e. at the centre of sample chamber) using the Fricke chemical dosimeter while alanine pellets, ethanol monochlorobenzene solution (ECB) and calibrated radiochromic film have been used for dose rate measurements in Seibersdorf, in Budapest and at NIST respectively. The dose per pulse delivered by the 12 MeV linear accelerator has been determined by means of the super Fricke chemical dosimeter and measurements have been regularly done before and after the irradiation of the devices. For the irradiation performed in Budapest with the LPR4 Tesla electron accelerator GAFChromic films, placed close to the transistors to be irradiated, have been used for dose measurements. Calibration of this film has been done using ethanol monochlorobenzene liquid dosimeter. Dosimetry using alanine pellets has been done at AERIAL. The irradiation doses were in the range from 50 Gy up to 10 kGy. All measured doses refer to dose in water.

2.4. Measurements of device characteristics

By knowing from previous studies [7, 8] that there is a linear correlation between the inverse carrier lifetime and the dose, expressed by the equation (1)

$$1/t_1 = 1/t_0 + kD \qquad (1)$$

where

t_1 is the postirradiation lifetime,

t_0 is the preirradiation lifetime,

D is the irradiation dose,

k is the radiation damage coefficient (in general k is substrate and irradiation condition dependent), our attention has been focused on the changes of carrier lifetime. For this purpose a portable instrument, whose schematic circuit diagram is shown in Fig. 1, was realized. It allows to measure on the spot, soon after irradiation, a physical parameter T directly related to the charge carrier lifetime and defined by the following equation:

$$T = t \cdot \ln(Q_s / t \cdot I_b) \quad (2)$$

where

t is the charge carrier lifetime,

Q_s is the stored charge,

I_b is the turnon base current.

By properly selecting the right driving conditions V_{BB}, $R_{BB(2)}$, $R_{C(2)}$, V_{CC} and I_{B1}, T can be considered a function of the lifetime only. Readout is accomplished merely by pressing a button to enable a digital display of the measured T value.

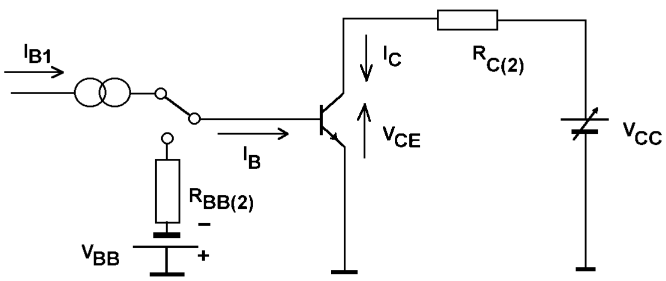


FIG. 1. Resistive load switching test circuit for measuring the parameter T. The driving conditions V_{BB}, $R_{BB(2)}$, $R_{C(2)}$, V_{CC}, I_{B1} have fixed values.

The deep levels introduced into the silicon structure of the transistor by g or electron irradiations have been also monitored by the deep level transient spectroscopy technique (DLTS) by using a commercial lockin type spectrometer. The DLTS technique, used to charecterize the traps in a semiconductor material, is based on measurements of timedependent capacitance signals. These are produced every time a quiescent reverse bias of a pn junction is changed by an appropriate bias pulse which injects carriers into the initially depleted region (filling pulse) [9]. As the previous bias conditions are restored, the population of carriers returns to its initial equilibrium, i.e. to the depletion condition, by emission of trapped carriers. This process is observed in form of a capacitance transient.

If single level centres are considered, the return to equilibrium is exponential, so it is possible to evaluate the emission rate by measuring the capacitance variations versus time, namely the difference in the capacitance values measured at two preselected time instants of the decay curve (rate window). DLTS spectra are obtained by scanning over a selected temperature range and represent capacitance differences as a function of the absolute temperature. As the temperature is varied, a peak is observed in the spectrum whenever the emission rate of a particular defect matches the selected rate window. The shape, the height and the position of these peaks along the temperature axis are directly related to the physical characteristics of the diefect introduced. The defect concentrations reported in Fig. 7 were obtained by using a modified ZothaWatanabe expression [10].

3. RESULTS

3.1. Irradiations

Batches of BULT118 transistors, supplied by SGS, have been irradiated up to doses of 5 kGy in different places and with different types of radiation sources. When the irradiation was done abroad, two of the transistors shipped were for control purposes. They were not irradiated and they always accompanied the other transistors except during irradiation. All that in order to take into account any possible change due to transport and storage. Measurements of the paramter T were done in Bologna. The changes of T with irradiation dose have been plotted as D(1/T) = 1/T 1/T_0 and are shown in Figs 2–4. These results indicate that the response of the transistor detectors is linear with absorbed dose up to 5 kGy, giving a linearity correlation coefficient r^2 = 0.9988 for 12 MeV electrons, r^2 = 0.9969 for 4 MeV electrons, r^2 = 0.995 for 2.5 MeV electrons and r^2 = 0.9909 for gamma irradiations. To check the stability of the lifetime changes produced by irradiation, the irradiated devices, together with some blanks, were left on the shelf of a cabinet without any special precaution. They have been measured over a year period. Some of the results are tabulated in Table III. The differences that have been found were randomly distributed and in the worst case not greater than 3% S.D. This is in agreement with the fact that transistors of the same type used for this study and electron irradiated, subjected to reliability tests ("high temperature reverse bias" HTRB) at 150°C for 1000 h, did not show any significant variations of the lifetime [11]. The results of these tests are reported in Fig. 5.

TABLE III. LONG TERM STABILITY OF TRANSISTORS

Sample	T_0 (ms)	Radiation	Dose	T (ms) date	T (ms) date
2	244	g ^{60}Co (FRAE) 24/03/95	50 Gy	226 (24/03/95)	225 (08/02/96)
3	239	//	//	236 //	234 //
37	227	//	500 Gy	198 //	201 //
49	233			231 //	228 //
194	230	e 12 MeV (FRAE) 24/05/95	2029 Gy	92 (24/05/95)	95 (08/02/96)
197	230	//	//	94 //	96 //
199	230			228 //	227 //
200	229			230 //	230 //

3.2. DLTS measurements

DLTS measurements have been carried out on the irradiated devices in order to characterize the recombination centres introduced by irradiation and affecting the carrier lifetime. The results of these measurements are reported in Figs 6 and 7. In Fig. 6 three peaks, labelled as E_1, E_2 and E_3, are clearly distinguishable. They correspond to the three main electron traps identified as the oxygenvacancy complex (A centre), the double negative $(VV)^=$ and single negative (VV) charge state of the divacancy, respectively. These three levels are all active recombination centres that can account for the changes of the lifetime, and consequently of the related electrical parameters, observed with irradiation [7]. Their thermal stability up to 150°C (the maximum working temperature of the devices), very important for safe operation of the device, is well kown from previous study [12] and the results, reported in Fig. 5, are a direct consequence of that.

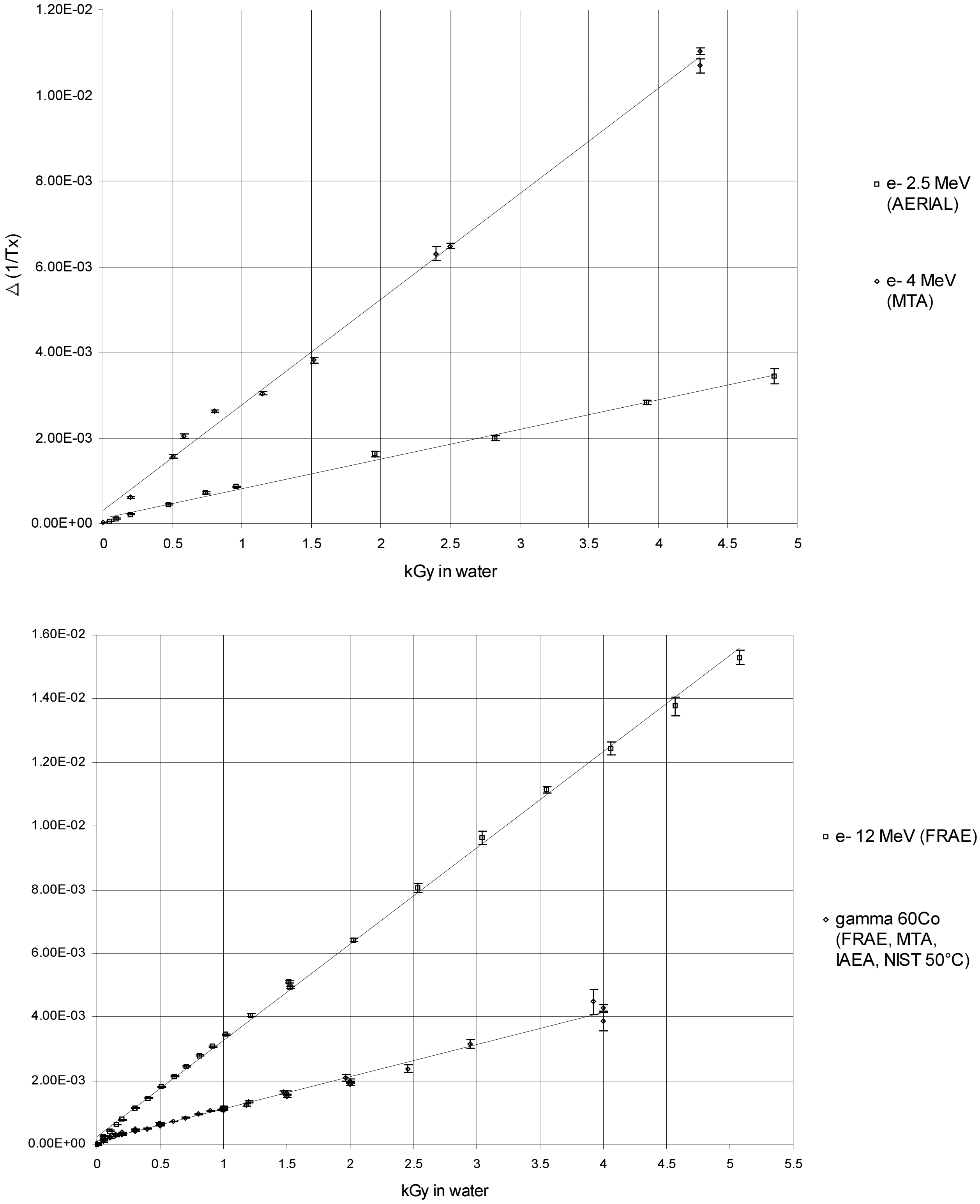


FIG. 2. Plot of $1/T - 1/T_0$ vs. dose for γ and electron irradiations performed in Bologna, Seibersdorf, Strasbourg, Budapest and Washington.

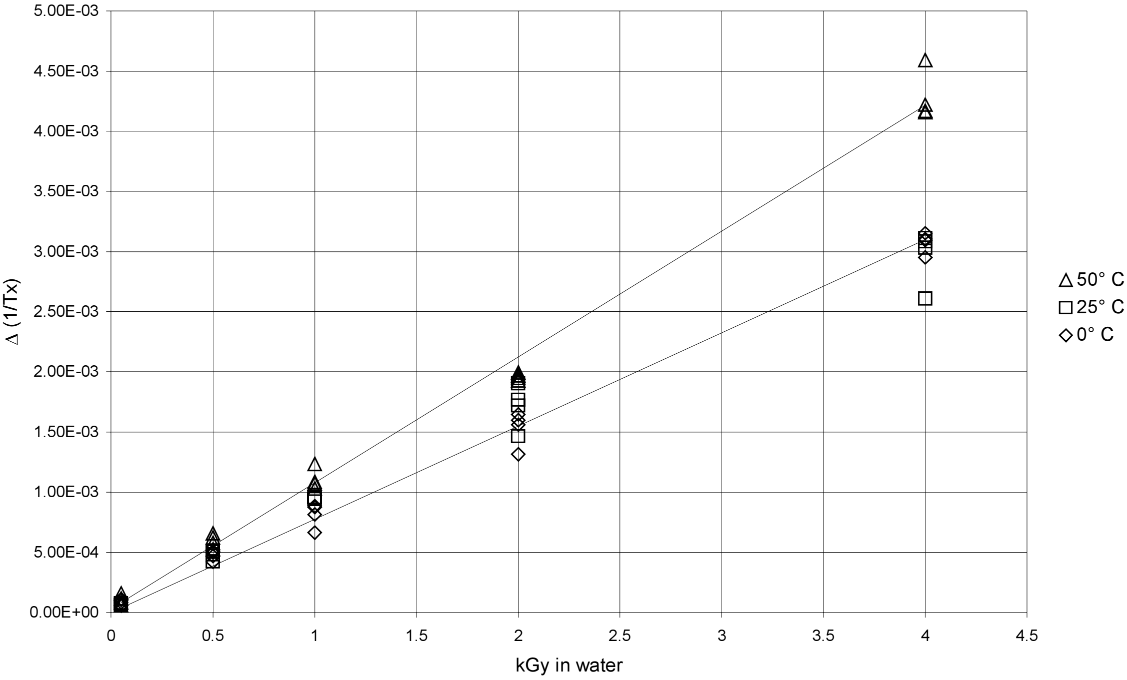


FIG. 3. Plot of 1/T 1/T$_0$ vs. dose for g irradiations performed at NIST, Washington, at different temperatures.

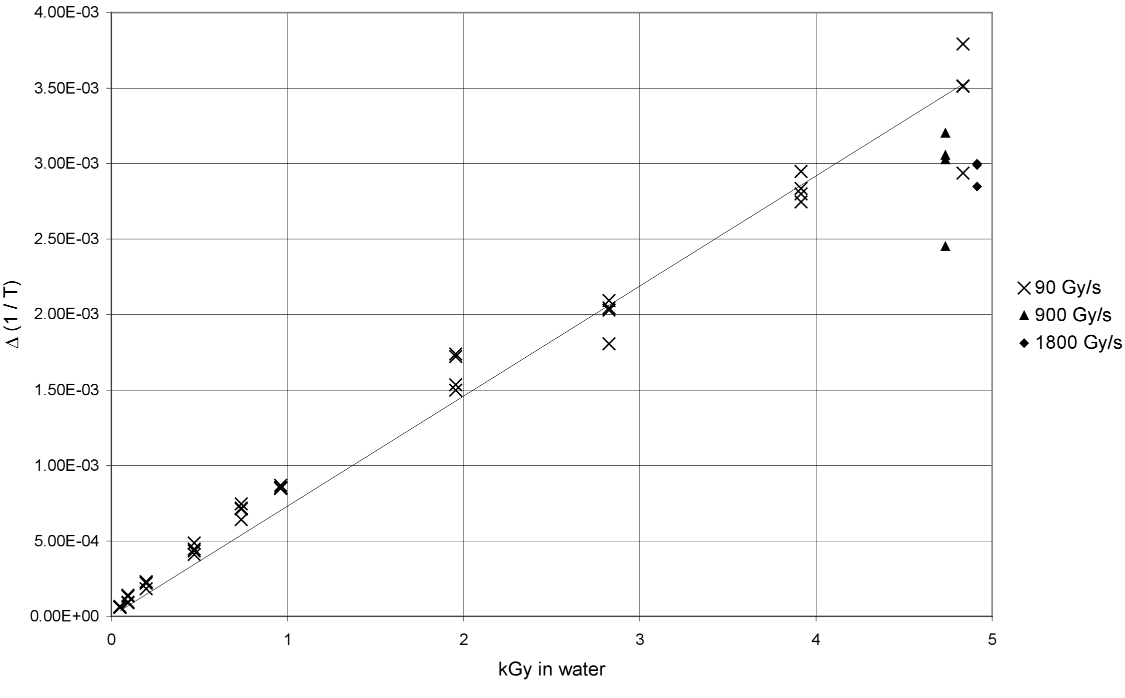


FIG. 4. Plot of 1/T 1/T$_0$ vs. dose for electron irradiations performed at AERIAL, Strasbourg, at different dose rates using a 2.5 MeV accelerator. The dose rates reported are mean dose rates over the whole scanned surface (24 cm by 6 cm).

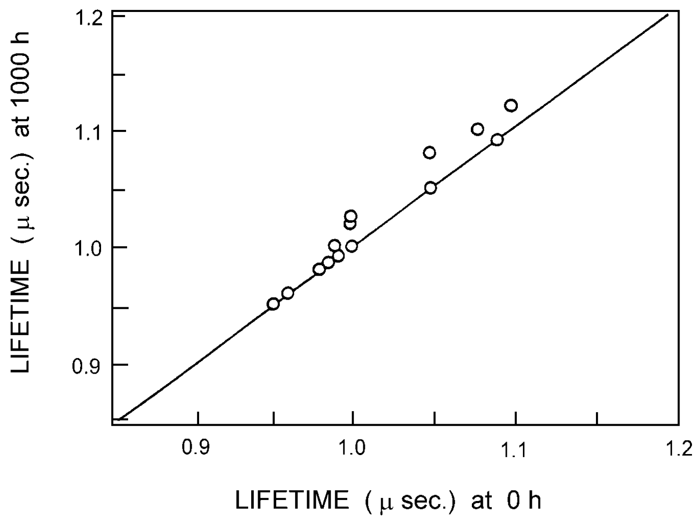


FIG. 5. Comparison of electron irradiated transistors after 1000 h annealing at 150ºC and soon after irradiation.

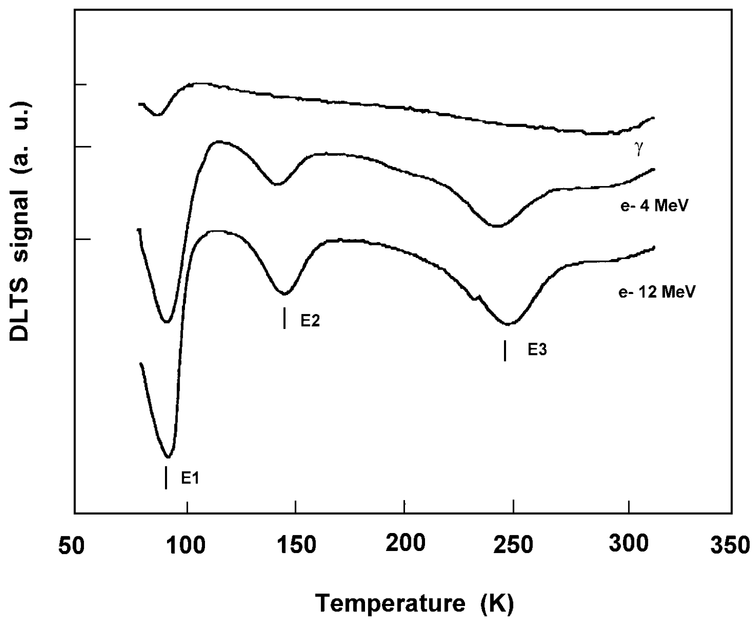


FIG. 6. DLTS spectra from transistors irradiated with g rays, 4 and 12 MeV electrons at a dose of 1.5 kGy. Rate window = 575 s[1].

4. CONCLUSIONS

The work on the characterization of these transistors as possible routine dosimeters is not yet completed. Only very few measurements have been done to evaluate the effect of different environmental conditions such as temperature and humidity. The results obtained from girradiations at controlled temperature of 0, 25 and 50°C show a temperature dependent behaviour of these devices (Fig. 3). The reason of that is not clear and more tests need to be done to confirm this effect. A large spread of lifetime values after irradiation has been observed in the low dose range (up to 150 Gy) for g rays, 2.5 and 4 MeV electrons, nevertheless the results obtained so far, are satisfactory and promising. The bipolar transistor's advantages are its small size, low cost, ease of use, good sensitivity, possibility of immediate reuse and its ability to record dose history (dose information is not lost during retrieval process). Moreover dose can be ascertained within minutes after irradiation with an inexpensive support equipment. All that bring us to conclude that such device may be suitable dosimeter for the daytoday monitoring of radiation process and its possible use should be taken into consideration. Particularly for 12 MeV electron irradiation the behaviour of the transistors is good; the spread of D(1/T) values is ≤ 10% for doses up to 100 Gy while it goes down to 34 % for the dose range 0.15–5 kGy. The irradiations done at different dose rates with the g sources and the few ones done using a 2.5 MeV accelerator indicate that dose rate does not seem to affect the response of the transistor (Fig. 4), at least in the explored range.

The silicon devices are made from a mixture of plastic, copper and silicon, and it is difficult to evaluate their radiation absorption characteristics. The small size of the devices relative to the range of the secondary electrons makes the mass collision stopping power the most important parameter, and the ratio of mass collision stopping powers is not changing much with energy, but below 12 MeV the ratios differ significantly. If the devices are calibrated and used in radiation fields with significant differences in radiation energy spectra, differences in responce may be expected.

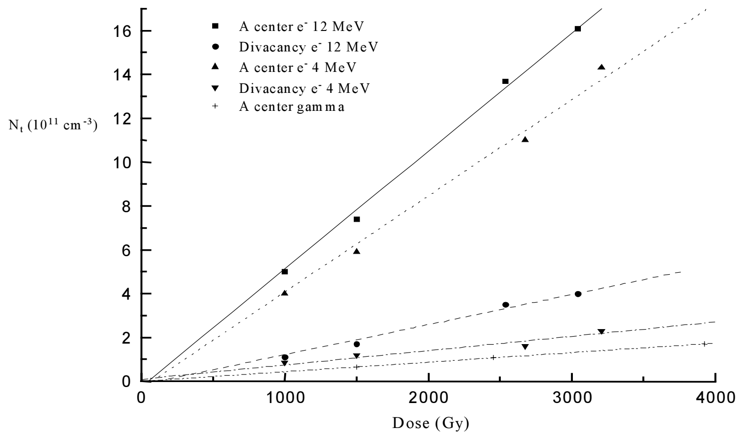


FIG. 7. Plot of concentration of A centre and divacancy vs. dose from irradiated transistors.

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REFERENCES

[1] RIKNER, G., GRUSELL, E., Phys. Med. Biol. **32** (1987) 1109 1107 and references therein.

[2] For more information see: a) SCHARF, K., Health Phys. **13** (1967) 575–586; b) MULLER, A.C., “The “n” on “p” solarcell doserate meter”, Manual on Radiation Dosimetry (HOLM N.W., BERRY R.J., Eds), Marcel Dekker, New York (1970) 423427; c) OSVAY, M., STENGER, V., FÖLDIAK, G., ”Silicon detectors for measurement ofhigh exposure rate gamma rays”, Biomedical Dosimetry, STI/PUB/401, IAEA, Vienna (1975) 623–632.

[3] MULLER, A.C., “The “p” on “n” solar cell integrating dosimeter”, Manual on Radiation Dosimetry (HOLM N.W., BERRY R.J., Eds), Marcel Dekker, New York (1970) 429–433.

[4] HARTSHORN, A., MACKAY, G., SPENDER, M., THOMSON, I., “Absorbed dose mapping in selfshielded irradiators using direct reading MOSFET dosimeters” (Proc. of the Health Physics Society), Annual Meeting, Boston, July 1995.

[5] GRÜNEWALD, T., RUDOLF, M., Food Irradiation Newsletter, FAO/IAEA **11** (1987) 42–47.

[6] EHLERMANN, D.A.E., “Dose distributions and methods for its determination in bulk particulate food materials”, Health Impact, Identification and Dosimetry of Irradiated Foods, Report of WHO Working Group (BÖGL, K.W., REGULLA, D.F., SUESS, M.J. Eds), ISHHeft 125, Institut fur Strahlenhygiene des Bundesgesundheitsamt, Neuherberg (1988) 415–419.

[7] FUOCHI, P.G., Radiat. Phys. Chem. **44** (1994) 431–440.

[8] BIELLEDASPET, D., Solid State Electron. **16** (1973) 1103–1123.

[9] LANG, D.V., J. Appl. Phys. **45** (1974) 3023–3032.

[10] GHEZZI, C., GOMBIA, E., VANZETTI, L., Material Science Forum Vol 1012 (1986) 1213–1218.

[11] ARCORIA, G., PATTI, A., FUOCHI, P.G., Radiat. Phys. Chem. **42** (1993) 1015–1018.

[12] BARBERIS, L., ICARDI, M., PORTESINE, M., TENCONI, S., DI MARCO, P.G., MARTELLI, A., FUOCHI, P.G., Radiat. Phys. Chem. **26** (1985) 165–172.

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