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.

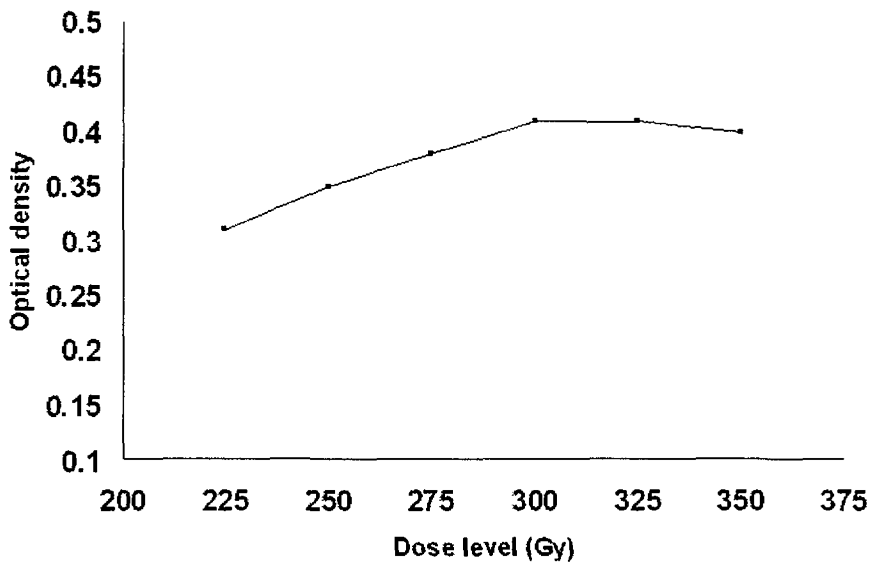

FIG. 1. Optical density vs dose for sterin 300.

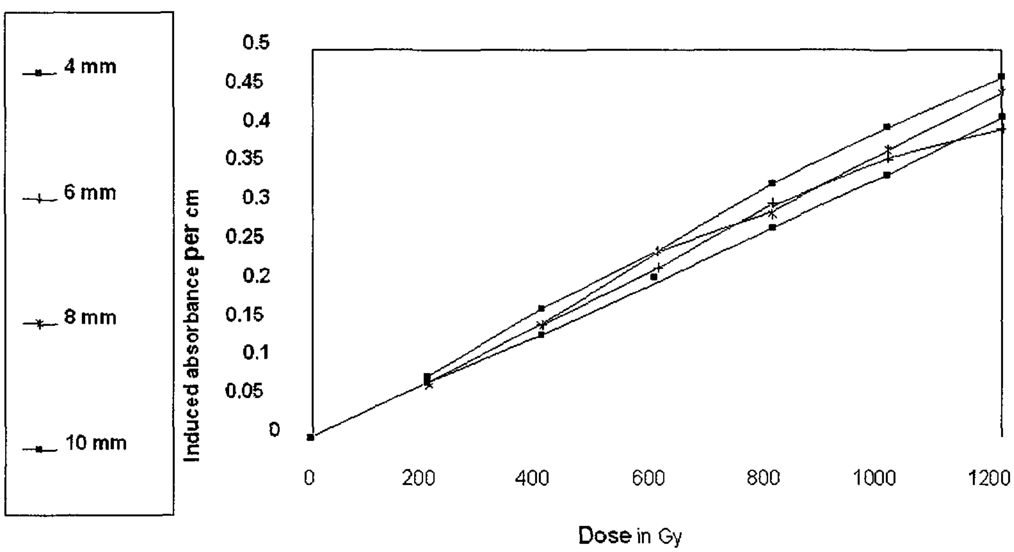

FIG. 2. Dose response of Pakistani clear PMMA.

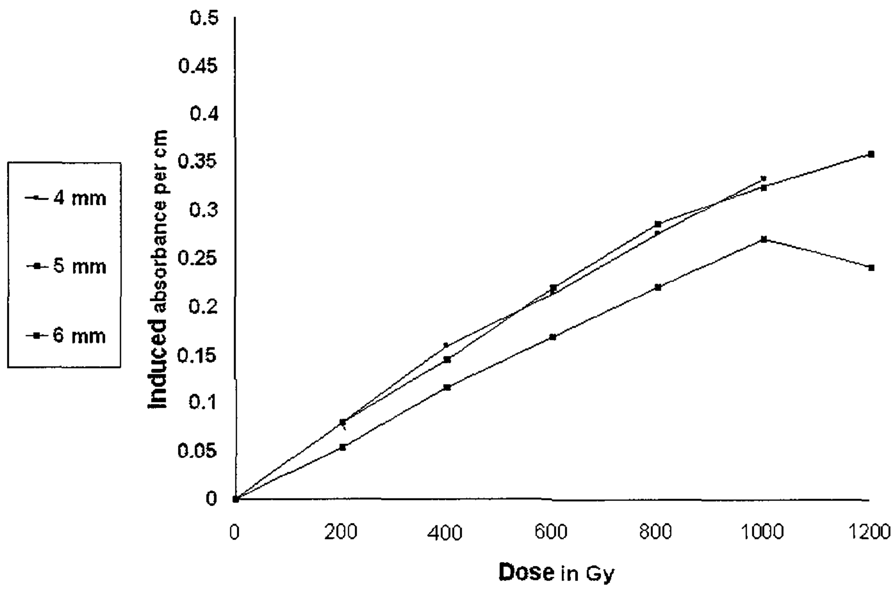

FIG. 3. Dose response of local clear PMMA.

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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