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			++	++

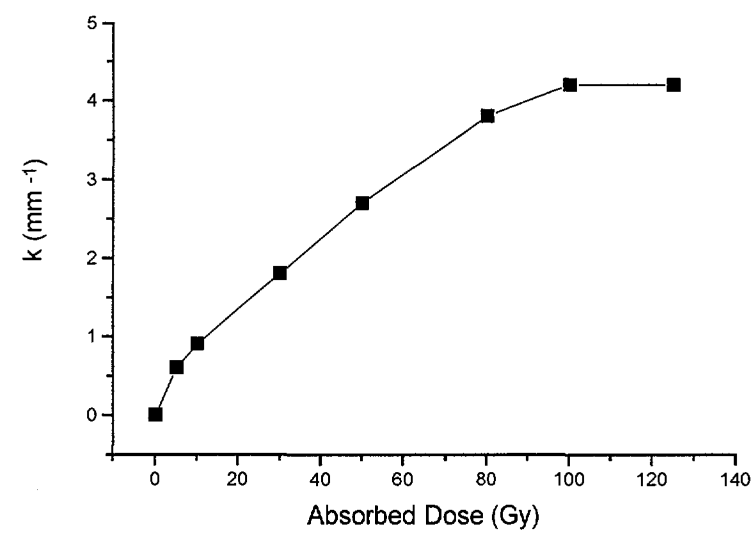

Figure 1. Dose response curve at 665 nm for 125 Gy INDICATOR irradiated with ^{60}Co gamma rays.

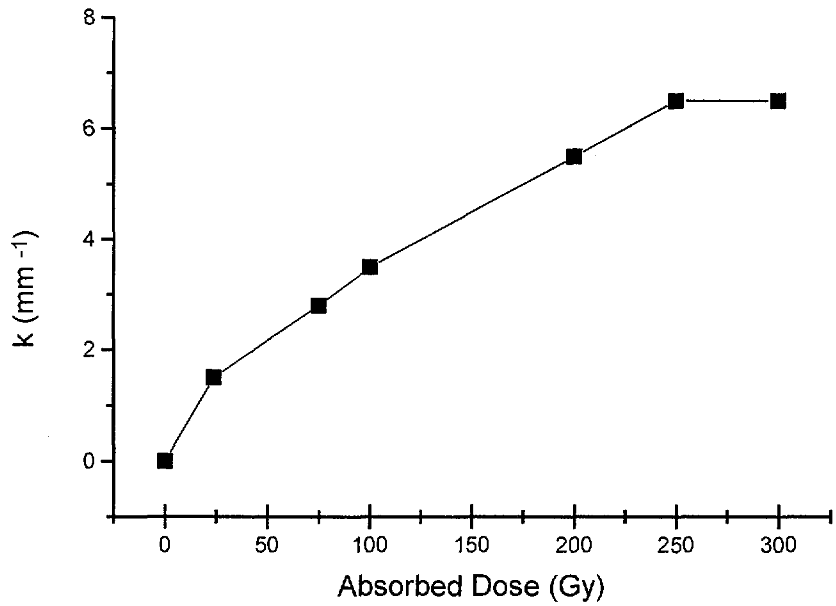

Figure 2. Dose response curve at 665 nm for 300 Gy INDICATOR irradiated with ^{60}Co gamma rays.

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			++	++

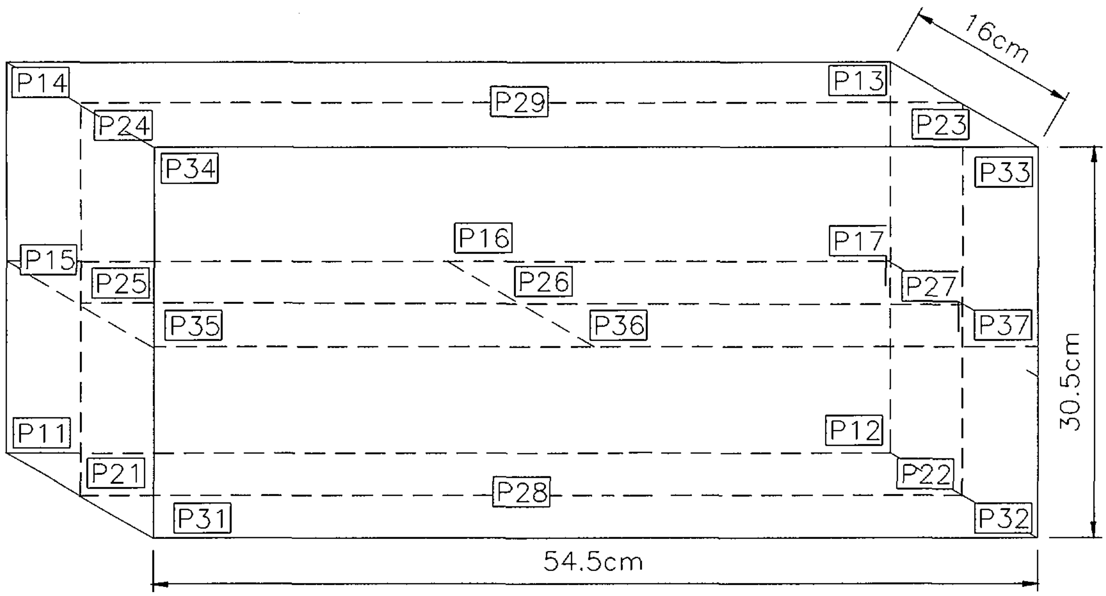

FIG. 3. Distribution of label dose indicators in wooden boxes.

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.

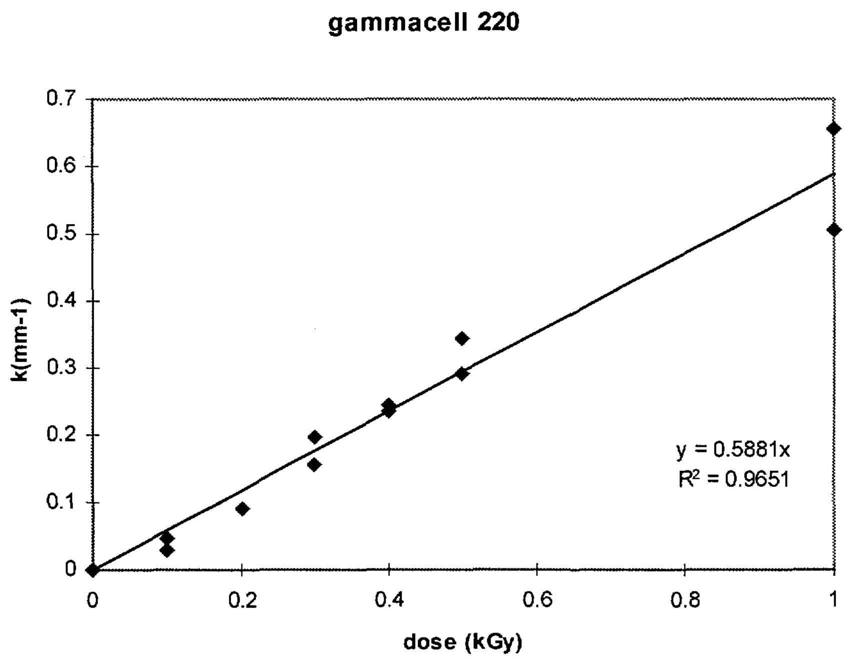

FIG. 4. Specific absorbance (530 nm) versus dose, Gammachrome YR, batch 5, Gammacell 220.

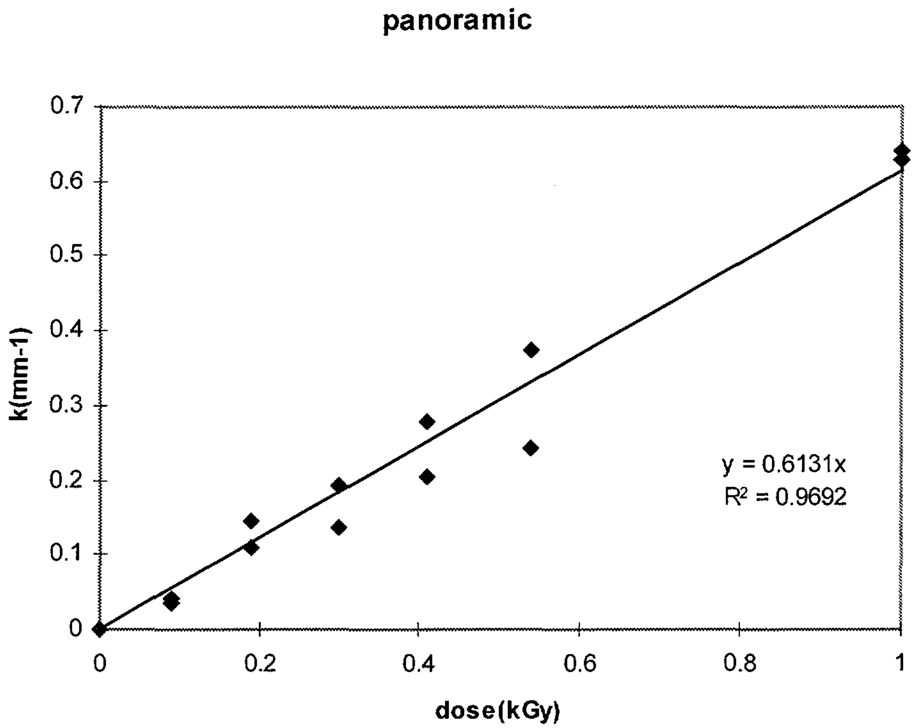

FIG. 5. Specific absorbance (530 nm) versus dose, Gammachrome YR, batch 5, panoramic ^{60}Co source.

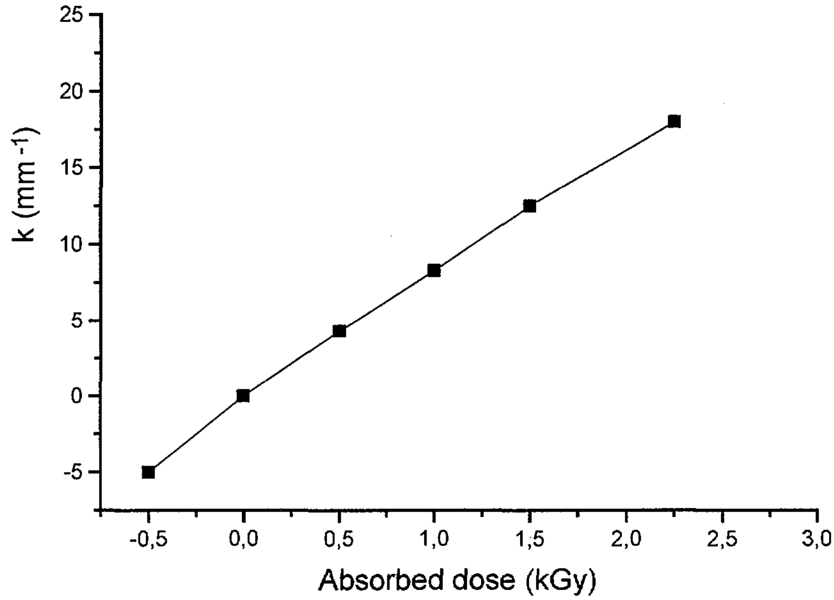

FIG. 6. Dose response curve for DM1260 films (spectrophotometric readings at 510 nm).

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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