DOSE MAPPING OF PRODUCTS WITH DIFFERENT DENSITY IRRADIATED IN ^{60}Co IRRADIATION FACILITY OF THE VINCA INSTITUTE, SERBIA

by

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Scientific paper
https://doi.org/10.2298/NTRP2001056R

The distribution of the absorbed dose within the irradiated product is a complex function of the product density and homogeneity, the position and shape of the radiation source, as well as the design of the irradiator. In this paper, detailed mapping of absorbed radiation doses in products of different density: gauze, plastic, and soil, is performed. Positions of minimum and maximum absorbed radiation dose were determined, and the homogeneity of irradiation of products was calculated using the ethanol-monochlorobenzene oscillotitrator dosimetry system.

Key words: gamma irradiation, dose mapping, ethanol-monochlorobenzene, ISO 11137

INTRODUCTION

The irradiation facility of the Vinca Institute of Nuclear Sciences was built with the assistance of the United Nations Development Fund (UNDP) and the International Atomic Energy Agency (IAEA) and put into operation in 1978. The radiation facility was designed by the Atomic Energy Commission and Conservatome of France for the activities of ^{60}Co sources up to 1 MCi. The ^{60}Co radiation source is plate-shaped. When the source is in a safe position, it is located at the bottom of the pool with water. In the operating position, the source is above the floor level of the irradiation cell. The products are irradiated in cycles of 328 boxes, which can be automatically transported and homogeneously irradiated using the conveyor system (ABP Company, France) and transport belts. Boxes must have standard dimensions: width 46 cm, length 46 cm, and height up to 43 cm. The maximum weight of the box can be 20 kg.

The boxes with products for irradiation are loaded into 82 cabins (vertically arranged by 4) that occupy three rows on each side of the source plane, which is lifted from the pool by a lifting mechanism to the operating position for irradiation. Cabins filled with product boxes are transported by conveyor mechanism from the warehouse for non-sterile goods through the maze to the radiation cell. Figure 1 shows the cabins in which are stored the products for irradiation and conveyor system.

The boxes are moved around the source and thus are irradiated. The moving of cabins with boxes through the cell is automated. The time a cabin remains in front of the radiation source is precisely determined. When the cabins leave the irradiation cell, the box level is automatically replaced. By turning and changing the level of the boxes after passing through the irradiation cell homogeneous irradiation of the boxes is achieved, as each side of the box is irradiated at each of the 4 passes. Upon completion of the irradiation process, the conveyor is emptied into the warehouse for sterile goods. The operation of the irradiation unit is controlled and managed from the control room.

Although the system designed in this way allows each box to receive an equal dose of radiation, this dose is not identical inside the box. Highly penetrating gamma rays do not irradiate the product equally on the surface and in the interior. The different homogeneity of the absorbed dose within the product also depends on the disposition of radiation sources (^{60}Co) within the frames.

Therefore, it is very important to determine the difference between the gamma radiation dose distribution within the product. Dose mapping is a way to determine if all the products in the irradiation process receive the required dose of radiation. By performing dose mapping, we can determine the dose distribution

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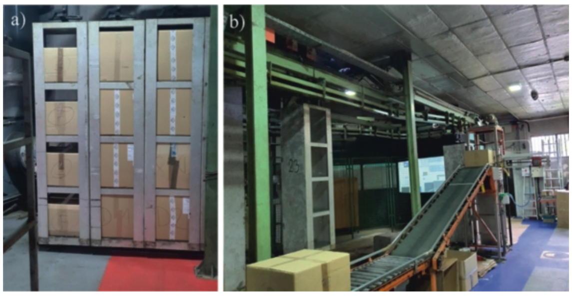

Figure 1. Irradiation cabins (a) conveyor system (b) irradiation facility of the Vinča Institute

within the product as well as the locations of the minimum and maximum absorbed radiation dose. Also, dose mapping is a requirement of the standard ISO 11137-1:2016 Sterilization of health care products - Radiation – Part 1: Requirements for development, validation and routine control of a sterilization process for medical devices [1].

Dose mapping in different irradiation facilities has been described in several papers [2-4]. However, due to the specific construction of the irradiator at the Vinča Institute, it is extremely important to perform dose mapping for products of different densities. This work was aimed at providing a complete dose mapping in irradiated products with different density (gauze, plastic, and soil), and at establishing positions of minimum and maximum absorbed radiation dose in different products.

MATERIALS AND METHODS

Synthesis of ethanol-monochlorobenzene

The ethanol-monochlorobenzene (ECB) dosimeter solution has been widely applied in gamma irradiations. The ECB dosimeter containing 24 vol.% of monochlorobenzene, 4 vol.% of distilled water, 0.04 vol.% of acetone, 0.04 vol.% of benzene, and 71.92 vol.% of ethanol was used as described in the literature [5].

High-purity chemicals (mandatory pro analysis) and triple distilled water quality are used to prepare the solution. All dishes used in the preparation of ECB dosimeters are washed with distilled water. In a glass container volume of 2 L, 480 ml of chlorobenzene, 80 ml of triple distilled water, 0.8 ml of acetone, 0.8 ml of benzene, and ethanol up to 2000 ml are poured [6]. The prepared solution is poured into 2 ml ampoules. The ampoules are previously selected by measuring the outer diameter in the range of 0.02 mm. The filled ampoules are clogged with the flame of the butane mixture and the oxygen.

Calibration of ECB dosimeters

To be sure of the accuracy of the results, a calibration must be performed for each lot of dosimeters [7, 8]. For each prepared lot of dosimeters, the 24 non-irradiated dosimeters were sent to the Reference laboratory, Riso High Dose Reference Laboratory – HDRL – DTU Nutech from Denmark, to be irradiated with different doses of radiation (5 kGy, 10 kGy, 15 kGy, 20 kGy, 25 kGy, and 35 kGy). Three different dosimeters were sent for each of the indicated doses. Based on these reference dosimeters, a calibration curve was constructed and used to calculate the absorbed dose from the irradiation process.

Placing dosimeters to perform dose mapping and gamma irradiation

The gamma irradiation was performed at the Radiation Facility of the Vinca Institute of Nuclear Sciences. The ECB dosimeters are placed in boxes in three planes for each of the products: front plane, center plane, and backplane relative to the position of the source. For lighter materials (gauze and plastic) we put nine dosimeters on each plane: at all 4 corners, at the midpoints of the edges and in the center, fig. 2. Boxes with lighter materials have a maximum height (43 cm) and are filled to the top.

For heavier materials (soil) the height of the box is 20 cm. So, we put 18 dosimeters, 6 dosimeters in each of three planes, fig. 3.

The ECB dosimeters are attached to the cardboard and inserted into boxes, fig. 4.

All boxes underwent a complete irradiation cycle (328 steps). Measuring equipment for ECB is the instrument OK-302/2 type oscillotitrator of Radelkis (Budapest, Hungary).

The dosimetry system in routine use at irradiation facilities is ECB dosimeter [6, 9]. Absorbed dose is determined through measurement results and a calibration curve. The calibration curve is based on the values obtained from the reference laboratory Riso High Dose Reference Laboratory (Denmark).

As the accuracy of the measurement on the oscillotitrator [10] is highly dependent on the mea-

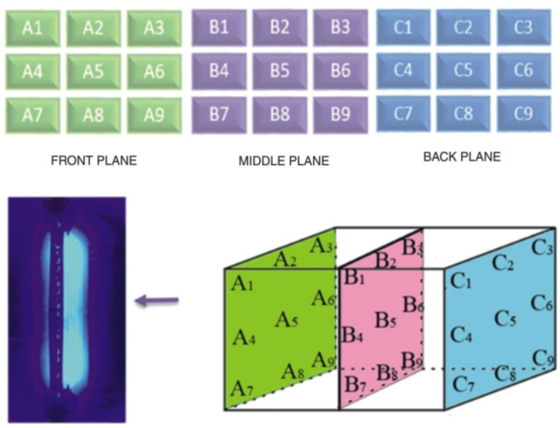

Figure 2. Arrangement of dosimeters by planes and arrangement of planes with dosimeters in the box and orientation towards the source for gauze and plastic

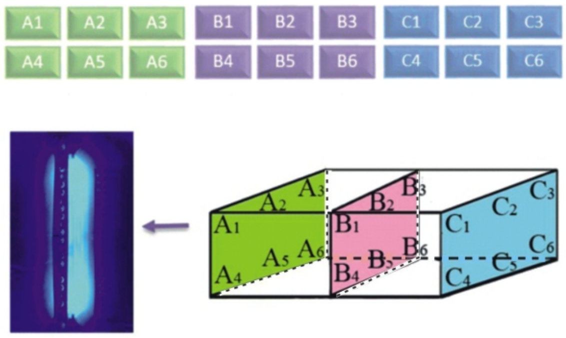

Figure 3. Arrangement of dosimeters by planes and arrangement of planes with dosimeters in the box and orientation towards the source for soil

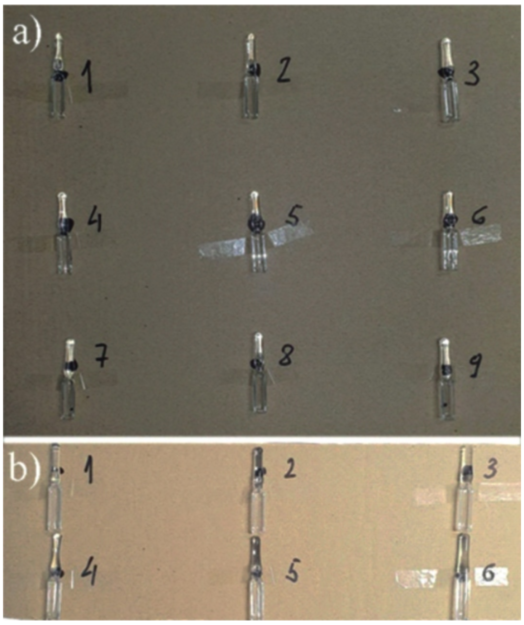

Figure 4. Arrangement of dosimeters on cardboard, for gauze and plastic (a) and soil (b)

surement temperature [11], all dosimeters are thermostatically controlled to a temperature of 20 °C, and all measurements are made at that temperature.

RESULTS AND DISCUSSIONS

Results of calibration

To form a calibration curve, we measured the values on the oscillotitrator scale, depending on the radiation dose to which the calibration dosimeters were exposed in the reference laboratory. The measurement is based on the determination of the electrical conductivity through the solution, and this value is shown on the scale as the signal strength, shown in arbitrary units, (a.u.).

Three different measurements were made. The following results were obtained, tab. 1.

Based on measured values, the calibration curve was performed, with 3rd polynomial fitting, fig. 5.

Based on this calibration curve and the obtained equation, all values of the absorbed radiation dose were calculated in this paper.

Dose mapping in the boxes filled with gauze

Boxes with gauzes are 43 cm high and filled to the top. Box has a volume of 0.091 m^3. The weight of the product without the box is 6 kg, so the average density of the product is 65.9 kgm^{-3}.

After irradiation and measurement of the value on the oscillotitrator, the following results were obtained, tab. 2.

Average dose: D_{av} = 29.42 kGy

Table 1. The values of the dosimeters irradiated in the reference laboratory, shown on the oscillotitrator

Dose	Number of ampule	The value on the oscilotitrator		
		1st measurement	2nd measurement	3rd measurement
5	D11	10.0	10.0	9.5
	D12	10.0	9.5	10.0
	D13	9.0	9.5	9.5
10	D21	29.0	29.5	29.0
	D22	30.0	30.0	29.5
	D23	29.0	29.0	29.0
15	D31	45.0	45.0	44.5
	D32	44.0	44.0	44.0
	D33	45.5	45.0	45.5
20	D41	60.0	60.0	60.0
	D42	59.0	59.0	60.0
	D43	59.0	59.5	59.0
25	D51	71.0	71.0	71.5
	D52	71.0	71.0	71.0
	D53	71.0	70.5	71.5
35	D61	90.0	89.5	90.0
	D62	90.0	90.0	89.5
	D63	89.5	90.0	90.0

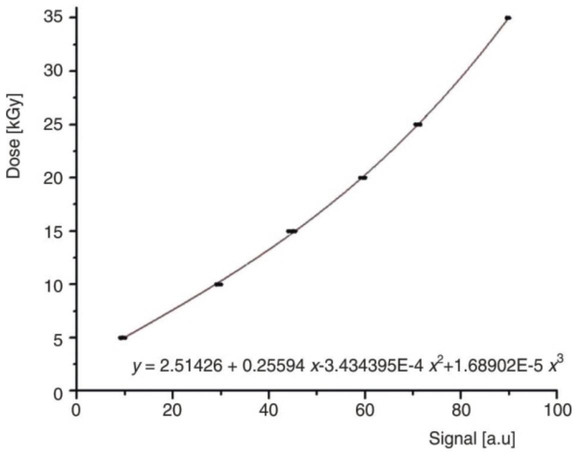

Figure 5. Calibration curve

To determine the measurement uncertainty, standard deviation and coefficient of variation were calculated. Based on these results, it follows:

Total overall deviation

$$Var_{\text{overall}} = \frac{\sum_{z=1}^{z_{\text{total}}} (n_z - 1)(SD)_z^2}{N - Z_{\text{total}}} = 0.062305\,\text{kGy}^2$$

where $n_Z = 3$ (measurement was done in 3 boxes), $N = 81$ (the total number of measurements), $Z_{\text{total}} = 27$ (number of measurement zones)

Relative uncertainty of measurement

$$\frac{\sqrt{Var_{\text{overall}}}}{D_{\text{sr}}} = 0.85\,\%$$

In the case of gauze products, the front and back planes receive a dose of 29.8 and 29.1 kGy, respectively, while the middle plane receives a dose of 25.8 kGy. The place of the minimum absorbed dose is at the middle of the bottom of the box, where an average value of 26.77 kGy was measured.

Homogeneity inside the box

$$U_{\text{av}} = \frac{D_{\text{max}}}{D_{\text{min}}} = \frac{31.40}{26.77} = 1.173$$

So, the maximum homogeneity in the box is about 17 %.

Dose mapping in boxes filled with plastic

Boxes with plastic material are 43 cm high and filled to the top. Box has a volume of 0.091 m^3. The weight of the product without the box is 15 kg, so the average density of the product is 164.8 kgm^{-3}.

After irradiation of the boxes with dosimeters, we measured the values on oscillotitrator, and based

Table 2. Dosimeters reading results for the box with gauze

Plane	Position	Calculated dose			Average dose [kGy]	Standard deviation $(SD)_Z$	Coefficient of variation (CV)	Comment
		Box 1 [kGy]	Box 2 [kGy]	Box 3 [kGy]				
FRONT	A1	31.2	30.7	30.7	30.87	0.24	0.76	
	A2	31.2	30.7	31.2	31.03	0.24	0.76	
	A3	29.9	29.7	29.9	29.83	0.09	0.32	
	A4	29.9	29.7	29.7	29.77	0.09	0.32	
	A5	**31.5**	**31.2**	**31.5**	**31.40**	**0.14**	**0.45**	**Maximum dose**
	A6	30.2	30.2	30.2	30.20	0.00	0.00	
	A7	29.2	28.9	29.2	29.10	0.14	0.49	
	A8	30.7	30.4	30.4	30.50	0.14	0.46	
	A9	30.2	29.9	29.9	30.00	0.14	0.47	
MIDDLE	B1	28.7	28.2	27.5	28.13	0.49	1.75	
	B2	28.2	27.5	28.2	27.97	0.33	1.18	
	B3	28.5	28.0	28.2	28.23	0.21	0.73	
	B4	27.7	27.7	27.7	27.70	0.00	0.00	
	B5	28.2	28.0	28.2	28.13	0.09	0.34	
	B6	27.7	27.7	27.5	27.63	0.09	0.34	
	B7	**26.5**	**27.3**	**26.5**	**26.77**	**0.38**	**1.41**	**Minimum dose**
	B8	27.3	27.0	27.0	27.10	0.14	0.52	
	B9	27.5	27.5	27.7	27.57	0.09	0.34	
BACK	C1	29.9	29.9	30.2	30.00	0.14	0.47	
	C2	31.7	31.5	31.5	31.57	0.09	0.30	
	C3	30.2	29.9	29.9	30.00	0.14	0.47	
	C4	29.9	29.9	29.9	29.90	0.00	0.00	
	C5	31.2	31.2	31.0	31.13	0.09	0.30	
	C6	31.0	30.7	29.9	30.53	0.46	1.52	
	C7	29.2	29.2	28.9	29.10	0.14	0.49	
	C8	30.2	30.2	31.5	30.63	0.61	2.00	
	C9	29.2	29.2	30.2	29.53	0.47	1.60	
	Minimum dose 26.77 kGy				Maximum dose 31.40 kGy			

Table 3. Dosimeters reading results for the box with plastic

Plane	Position	Calculated dose			Average dose [kGy]	Standard deviation $(SD)_Z$	Coefficient of variation (CV)	Comment
		Box 1 [kGy]	Box 2 [kGy]	Box 3 [kGy]				
FRONT	A1	30.9	30.3	30.3	30.50	0.28	0.93	
	A2	30.9	30.3	30.9	30.70	0.28	0.92	
	A3	30.3	30.3	30.9	30.50	0.28	0.93	
	A4	29.3	29.3	29.8	29.47	0.24	0.80	
	A5	**31.1**	**31.4**	**31.4**	**31.30**	**0.24**	**0.78**	**Maximum dose**
	A6	29.3	29.3	30.3	29.63	0.47	1.59	
	A7	28.5	28.8	28.8	28.70	0.14	0.49	
	A8	29.3	28.8	28.8	28.97	0.24	0.81	
	A9	28.5	28.8	28.8	28.70	0.14	0.49	
MIDDLE	B1	27.0	28.0	27.3	27.43	0.42	1.53	
	B2	27.8	27.3	27.3	27.47	0.24	0.86	
	B3	27.8	27.5	27.5	27.60	0.14	0.51	
	B4	26.8	26.8	26.1	26.57	0.33	1.24	
	B5	27.8	26.8	26.8	27.13	0.47	1.74	
	B6	27.3	26.8	27.3	27.13	0.24	0.87	
	B7	**25.8**	**25.4**	**25.8**	**25.67**	**0.19**	**0.73**	**Minimum dose**
	B8	25.8	26.1	25.8	25.90	0.14	0.55	
	B9	26.1	25.6	26.1	25.93	0.24	0.91	
BACK	C1	29.6	29.8	29.8	29.73	0.09	0.32	
	C2	30.6	30.6	30.3	30.50	0.14	0.46	
	C3	29.8	30.3	30.3	30.13	0.24	0.78	
	C4	29.3	28.8	28.8	28.97	0.24	0.81	
	C5	29.3	29.8	29.3	29.47	0.24	0.80	
	C6	29.0	28.8	28.8	28.87	0.09	0.33	
	C7	28.5	27.3	27.8	27.87	0.49	1.77	
	C8	28.8	28.8	28.3	28.63	0.24	0.82	
	C9	28.5	27.3	27.8	27.87	0.49	1.77	
	Minimum dose – 25.67 kGy				Maximum dose – 31.30 kGy			

on the calibration curve we calculated the received irradiation dose of gamma radiation. The results of dose measurements in gauze boxes are shown in tab. 3. We calculated the average dose, standard deviation and coefficient of variation.

Average dose: D_{av} = 28.57 kGy

To determine the measurement uncertainty, standard deviation and coefficient of variation were calculated. Based on these results, it follows

Total overall deviation

$$Var_{overall} = \frac{\sum_{z=1}^{z_{total}} (n_z - 1)(SD)_z^2}{N - Z_{total}} = 0.080247\ \text{kGy}^2$$

where n_Z = 3 (measurement was done in 3 boxes), N = = 81 (the total number of measurements), Z_{total} = 27 (number of measurement zones)

Relative uncertainty of measurement

$$\frac{\sqrt{Var_{overall}}}{D_{sr}} = 0.99\ \%$$

From the results, the front and back planes receive an average dose of 30.3 kGy and the middle plate an average dose of 27.7 kGy. The minimum and maximum absorbed dose positions are identical to those of boxes with gauze.

Homogeneity inside the box

$$U_{av} = \frac{D_{max}}{D_{min}} = \frac{31.40}{25.67} = 1.219$$

So, the maximum homogeneity in the box is about 22 %.

Dose mapping in the boxes filled with soil

Boxes with soil are 20 cm high and filled to the top. Box has a volume of 0.042 m^3. The weight of the product without the box is 20 kg, so the average density of the product is 476.2 kgm^{-3}. It should be noted that this is the average density of the product in the box, since the soil is packed in smaller bags, so it does not take up the complete volume of the box. After irradiation and measurement of the value on the oscillotitrator, the following results were obtained, tab. 4.

Average dose: D_{av} = 25.40 kGy

Total overall deviation

$$Var_{overall} = \frac{\sum_{z=1}^{z_{total}} (n_z - 1)(SD)_z^2}{N - Z_{total}} = 0.037037\ \text{kGy}^2$$

Table 4. Dosimeters reading results for the box with soil

Plane	Position	Calculated dose			Average dose [kGy]	Standard deviation $(SD)_Z$	Coefficient of variation (CV)	Comment
		Box 1 [kGy]	Box 2 [kGy]	Box 3 [kGy]				
FRONT	A1	27.0	27.2	27.0	27.07	0.12	0.43	
	A2	27.2	27.0	27.2	27.13	0.12	0.43	
	A3	27.4	27.2	27.4	27.33	0.12	0.42	
	A4	26.5	26.5	26.5	26.50	0.00	0.00	
	A5	26.5	26.5	27.0	26.67	0.29	1.08	
	A6	26.5	26.5	26.5	26.50	0.00	0.00	
MIDDLE	B1	22.5	22.2	22.5	22.40	0.17	0.77	
	B2	**20.8**	**20.8**	**21.2**	**20.93**	**0.23**	**1.10**	**Minimum dose**
	B3	22.9	22.5	22.9	22.77	0.23	1.01	
	B4	22.5	22.5	22.5	22.50	0.00	0.00	
	B5	21.2	21.0	21.2	21.13	0.12	0.55	
	B6	21.8	21.2	21.2	21.40	0.35	1.62	
BACK	C1	27.4	27.0	27.4	27.27	0.23	0.85	
	C2	27.0	27.0	27.0	27.00	0.00	0.00	
	C3	27.9	27.4	27.9	27.73	0.29	1.04	
	C4	27.4	27.4	27.4	27.40	0.00	0.00	
	C5	**28.4**	**28.4**	**27.9**	**28.23**	**0.29**	**1.02**	**Maximum dose**
	C6	27.0	27.4	27.4	27.27	0.23	0.85	
	Minimum dose – 20.9 kGy				Maximum dose – 28.2 kGy			

where $n_Z = 3$ (measurement was done in 3 boxes), $N = 54$ (the total number of measurements), $Z_{total} = 18$ (number of measurement zones)

Relative uncertainty of measurement

$$\frac{\sqrt{Var_{overall}}}{D_{sr}} \quad 0.76\ \%$$

In the case of soil, the front and back planes receive a dose of 26.9 and 27.4 kGy respectively, while the middle plane receives a dose of 21.8 kGy. The minimum dose place is at the top of the middle plane.

Homogeneity inside the box

$$U_{av} \quad \frac{D_{max}}{D_{min}} \quad \frac{28.23}{20.93} \quad 1.348$$

So, the maximum homogeneity in the box is about 35 %.

Dependence of the absorbed radiation dose as a function of product density

Based on the obtained data, a diagram of the dependence of the absorbed radiation dose on the density of the irradiated product was constructed, fig. 6.

From fig. 6 one can see that the irradiation dose absorbed in the material decreases almost linearly with increasing density of the material. Similar results have been described in the literature [12, 13]. Based on the latter, it is easy to estimate the required irradiation time for materials of different densities.

Also, we graphically presented the change in the homogeneity of the absorbed dose within the box, depending on the density of the material, fig. 7.

It can be concluded that the homogeneity of the absorbed dose increases with increasing product den-

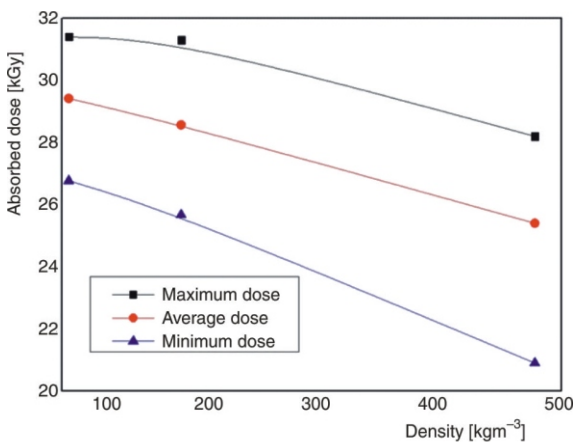

Figure 6. Dependence of the absorbed radiation dose as a function of product density

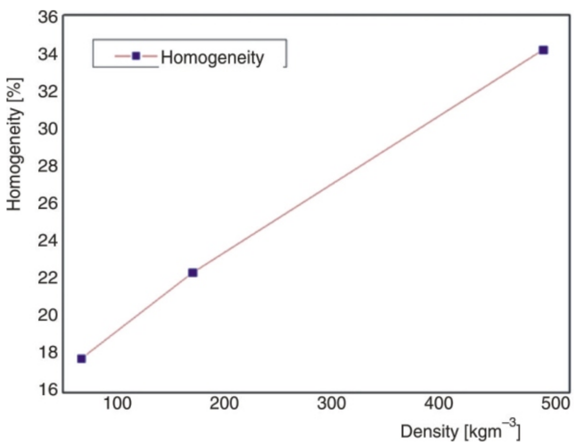

Figure 7. Dependence of the dose homogeneity as a function of product density

sity [14]. Less homogeneity means that the difference between the minimum and maximum absorbed dose is smaller. Thus, for a lower density of products, the absorbed radiation dose is more properly distributed within the product.

CONCLUSIONS

Dose mapping is a method that allows us to determine the distribution of doses within the irradiated product. It is very important to determine the minimum and maximum radiation dose within the product, as it is the responsibility of the irradiator to irradiate each part of the product with the minimum required dose. Also, the upper limit of the delivered dose, specified by the user, must not be exceeded.

From the data analyzed in this paper, it can be concluded that the density of the product significantly affects the radiation dose that the product will absorb. We have concluded that the irradiation dose absorbed in the material decreases almost linearly with increasing density of the material. Also, the homogeneity of the absorbed dose increases with increasing product density. For the lower density of products, the absorbed radiation dose is more properly distributed within the product.

ACKNOWLEDGMENT

Authors acknowledge the financial support of the IAEA (Project RER1019 – Enhancing Standardized Radiation Technologies and Quality Control Procedures for Human Health, Safety, Cleaner Environment and Advanced Materials)

AUTHORS' CONTRIBUTIONS

All measurements were performed by B. M. Ranković and N. R. Nikolić. The measurement setup was conceived and prepared by S. B. Mašić. The theoretical analysis was carried out by I. T. Vujčić. All authors analyzed and discussed the results and reviewed the manuscript.

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Received on January 31, 2020
Accepted on June 12, 2020

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МАПИРАЊЕ ДОЗЕ ЗРАЧЕЊА ЗА ПРОИЗВОДЕ РАЗЛИЧИТИХ ГУСТИНА ОЗРАЧЕНЕ НА ^{60}Co РАДИЈАЦИОНОМ ПОСТРОЈЕЊУ ИНСТИТУТА ВИНЧА, СРБИЈА

Расподела апсорбоване дозе унутар озраченог производа представља сложену функцију густине и хомогености производа, положаја и облика извора зрачења, као и дизајна постројења за озрачивање. У овом раду извршено је детаљно мапирање доза апсорбоване радијације у производима различите густине: газе, пластике и земље. Одређене су позиције минималне и максималне апсорбоване дозе зрачења, а хомогеност зрачења производа је израчуната помоћу дозиметријског система ECB/осцилотитратора.

Кључне речи: гама озрачивање, мапирање дозе, ECB, ISO 11137