ACCURACY IN DETERMINING ABSORBED IRRADIATION DOSE AT DIFFERENT TEMPERATURE MEASUREMENTS USING ETHANOL-CHLOROBENZENE Oscillotitrator System

by

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Ethanol-chlorobenzene/oscillotitrator dosimetry system is widely used in controlling the irradiation process in gamma facilities. The ethanol-chlorobenzene dosimetry system provides a reliable means of measuring absorbed dose. It is based on a process of radiolytic formation of hydrochloric acid in aqueous ethanolic solutions of chlorobenzene by ionizing radiation. The irradiation temperature dependence of dosimeter response is a complex function of dose and temperature for each concentration of chlorobenzene. At different temperature the mobility of conducting species from hydrochloric acid is changed leading to different oscillotitrator deflections during high-frequency conductometric readout. In this paper, we examined the influence of temperature on the calculation of the radiation dose. We showed that the temperature significantly influenced the measurement results, and that the calibration curve has to be formed at the irradiation temperature in order to obtain precise values of the absorbed dose.

Key words: ethanol-chlorobenzene, oscillotitrator, dosimetry, radiation

INTRODUCTION

Industrial application of radiation technology is expanding, especially in the sterilization of medical devices and food irradiation. For this purpose, traditional radiation sources such as ^{60}Co and ^{137}Cs are used. Lately, high power electron accelerators are coming to the market. New standards and regulations related to healthcare products and food treatment are continuously being introduced by international and national authorities. There are several standards developed by the regional and international bodies, such as the International Organization for Standardization (ISO), the European Committee for Standardization (CEN), and the American Society for Testing and Materials (ASTM) which provide recommendations and guidelines for the radiation processes. The most important requirements of those standards are process control and process validation. For those protocols, dosimetry is essential [1].

Dosimetry is the measurement of absorbed ionizing radiation doses [2]. It describes the relationship between the absorbed dose in a dosimeter and in the surrounding material. The dosimeter may be any material to which a certain property changes under the influence of radiation. The main requirements for the selection of materials to be used as a dosimeter are that the changing property is measurable, the changes are relatively stable over time, and that the changes are reproducible. A dosimeter system consists of three components: physical or chemical dosimeters, the instrument for measuring the relevant radiation-induced effect in the dosimeter and the procedure for use [3].

Several dosimetry systems are being used for radiation processing. One of the most commonly used dosimeter systems in gamma irradiation facilities is the ethanol-chlorobenzene (ECB) – oscillotitrator system. The ECB dosimetry system provides a reliable means of measuring dose based on the radiolytic formation of hydrochloric acid (HCl) in the aqueous ethanolic solution of chlorobenzene by ionizing radiation [4, 5]. Determining the content of the produced HCl can be done in three ways: by titration to deter-

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mine the concentration of chloride ions, by spectrophotometric readout at 485 nm, and by high-frequency measurement of change in the dielectric constant which can be undertaken with the solution still in its sealed ampoule [6]. Last measurement method uses an oscillotitrator as an instrument for measurement of change in the conductivity. In that case, dosimeters can be read up to about 3 years [7], and according to some experiences even after more than 20 years. Another advantage of using this method in comparison to chemical titration is that when using the oscillotitrator, the measured ampoules are closed, so the contact between the person who measures with ECB solution is avoided. Also, the advantage is based on the response which is instantaneous, and the electrodes need not be in direct contact with the solution. This is especially important because the ECB solution is toxic. Also, it is easier to manipulate with closed ampoules during the radiation process.

It is well known that the temperature change does not have an effect on the concentration of formed HCL [8] when the concentration is measured by chemical titration. However, when oscillotrator is used for measurement, a change in the values on the scale is observed depending on ampoule temperature. This is very important to consider because, during the process of goods irradiation, there may occur a change of temperature in a warehouse or bunker due to different weather conditions, as well as due to heating in processes in plants with high source activity. Further, the ambient temperature in the dosimetry laboratory can vary depending on the season, the outside temperature, the way of heating and cooling the room, *etc*. Some dosimetry laboratories achieve the thermostatic conditions by holding the dosimeters at the ambient temperature of the measuring room for a period of one hour in order to equalize the temperature of the measured and the reference dosimeters. However, it is often necessary to perform dosimetric measurements much faster, due to the requests of users who come to take over their treated goods. The aim of this paper is to prove that thermostatic treatment is a necessary part of determining the dose of absorbed radiation using the ECB-oscillotitrator system in the radiation processing units. This knowledge leads to a more precise determination of the dose of absorbed radiation in the sterilization of medical equipment, health care products, food conservation, sterilization of pharmaceuticals and cosmetics, treatment of soil, cultural heritage and all other products that can be irradiated in radiation facilities.

First, we performed measurements of ampules irradiated in the reference laboratory *Riso High Dose Reference Laborator* with known doses, and then we measured the doses at the ampoules from the standard irradiation batch, using calibration at room temperature and calibration at ampoule temperatures. We concluded that the temperature of the ampoule significantly influences the measurement results and that the measurement error decreases if the calibration is performed at the temperature of the ampoules.

MATERIALS AND METHODS

Preparation of ethanol-chlorobenzene

The dosimetric solution is a 24 % solution of chlorobenzene in ethanol. High purity chemicals (mandatory *pro analysis*) and triple- distilled water quality (milli-q water system) are used to prepare the solution. All dishes used in the preparation of ECB dosimeters are washed with distilled water quality for radiochemical tests. In a glass container volume of 2 liters, 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. 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 [9].

To maintain the constant temperature of the EBC ampoule, we used a hand-made thermostatic chamber. This chamber has a system of heaters that quickly heat up ampules to the required temperature, as well as a cooling system, should it be necessary to lower the temperature of the ampoule. It can reach temperatures from 5 °C to 60 °C.

Instruments and measurements

Measurements were made by oscillotitrator OK-302/1 supplied by Radeliks Electrochemical Instruments, Budapest. The oscillotitrator consists of the reader including controls and the oscillator, which is built together with the ampoule holder. Ampoules are placed in a holder and value is read on the scale. The oscillotitrator measurement is based on determination of electrical conductivity through various solutions. With an increase in the radiation dose, they are exposed to, the solution increases the conductivity due to the radiolytic formation of HCl, and this is manifested by the higher value of the signal (shown in arbitrary units, a.u.) on the scale. Instrument calibration is performed by reference dosimeters irradiated in the reference Riso Laboratory with the required doses of radiation. For each lot of dosimeters, the 24 non-irradiated dosimeters are sent to the *Riso Laboratory,* and it is required that they be irradiated with different doses of radiation. (1 kGy, 3 kGy, 5 kGy, 10 kGy, 15 kGy, 20 kGy, 30 kGy, and 55 kGy). Three different dosimeters are sent for each of the indicated doses. Based on the reference dosimeters a calibration curve is constructed and used to calculate the radiation dose from the irradiation process. The average oscillometric measurement value is determined based on the difference in the measurement results of the three ampoules irradiated with the same dose, as stated.

The formation of the calibration curve is routinely performed at room temperature, and in this paper, we will show that the calibration should be performed at the temperature at which the routine measurements are performed. Routine and reference dosimeters should always be thermostated to the temperature of the calibration curve, in order to avoid measurement errors.

RESULTS AND DISCUSSION

To determine the dependence of the temperature on the response of the oscillotitrator, dosimeters irradiated in the *Riso Laboratory* with precise doses of 5, 10, 15, 20, 25, and 35 kGy were used. We used three dosimeters for each of the above doses.

Dosimeters are exposed to temperatures of 5, 10, 15, 20, 30, 40, and 50.

Table 1 shows the results of the measurements on the oscillotitrator. In order to avoid a subsequent change in temperature after thermostat, and before measuring, the ampoules are placed directly from the thermostat into the oscillotitrator holder. In the columns, the average measurement value of three different dosimeters is given for each individual dose and measurement temperature.

From tab. 1 one can see that the oscillotitrator response is significantly higher with the temperature increase. Based on these responses and known dose values, curves of addiction have been constructed, fig. 1.

When those values are converted to the third-degree polynomials, equations that describe the dependence of the dose and the oscillotitrator signal for each individual temperature are obtained

5 °C

$$Y = 3.62422 + 0.35205\,X - 0.00193\,X^2 + 4.21452\cdot10^{-5}\,X^3 \tag{1}$$

10 °C

$$Y = 2.93609 + 0.33515\,X - 7.10327\cdot10^{-4}\,X^2 + 1.92086\cdot10^{-5}\,X^3 \tag{2}$$

15 °C

$$Y = 3.18957 + 0.2298\,X + 0.00102\,X^2 + 6.10497\cdot10^{-6}\,X^3 \tag{3}$$

20 °C

$$Y = 2.74126 + 0.22054\,X + 7.179364\cdot10^{-4}\,X^2 + 8.28564\cdot10^{-6}\,X^3 \tag{4}$$

30 °C

$$Y = 2.64315 + 0.23435\,X - 1.83072\cdot10^{-4}\,X^2 + 1.18522\cdot10^{-5}\,X^3 \tag{5}$$

40 °C

$$Y = 0.57988 + 0.42039\,X - 0.00569\,X^2 + 4.92729\cdot10^{-5}\,X^3 \tag{6}$$

50 °C

$$Y = 1.66345 + 0.24602\,X - 0.00189\,X^2 + 2.04496\cdot10^{-5}\,X^3 \tag{7}$$

After having determined the equations, we selected ten dosimeters irradiated with an unknown radiation dose. Those were dosimeters for controlling the process of industrial sterilization of medical devices (required dose of minimum 25 kGy) and food conservation (required dose of minimum 5 kGy). Five randomly selected dosimeters from each dose were used, and measured after being thermostated at temperatures of 5 °C, 10 °C, 20 °C, 30 °C, 40 °C, and 50 °C.

We calculated the absorbed dose in two ways: using the calibration done at the room temperature (20°C), and the calibration performed at the temperature at which the dosimeters from the process were ex-

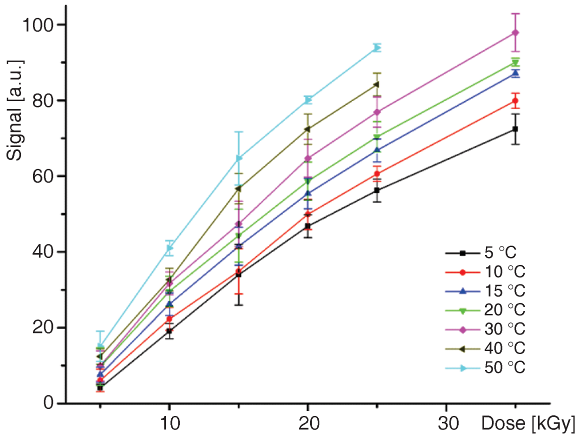

Figure 1. The ratio of the oscillotitrator signal and the dose depending on the ampoule temperature

Table 1. The value of the signal response measured on the oscillotitrator at different temperatures of the ampoule with the ECB dosimeter

Dose	Temperature						
	5 °C	10 °C	15° C	20° C	30° C	40 °C	50°C
	Signal on the oscillotitrator (a. u.)						
5 kGy	4.1	6.1	7.6	9.7	9.9	12.4	15.1
10 kGy	19.1	22.3	26.2	29.6	31.7	32.7	41.0
15 kGy	34.0	34.9	41.5	44.3	47.4	56.7	64.7
20 kGy	46.8	49.9	55.4	58.7	64.7	72.4	80.1
25 kGy	56.2	60.6	66.8	70.4	76.9	84.2	93.9
35 kGy	72.4	79.9	87.1	90.1	97.9	Value outside the range	

posed, by using the previous equations. The aim of two different measurements was to determine the amount of error in the measurement at room temperature without taking calibration into consideration. The following results were obtained, tab. 2. The table shows the average values of dose measurement with the room temperature calibration, the calibration of the ampule temperature, as well as the expected dose of measurement. An error in dose measurement using calibration at room temperature in comparison to the measurement of the dose by using calibration at ampoule temperature was obtained.

From tab. 2 one can see that the measurement errors increase with the increase of the temperature differences relative to room temperature. Thus the deviations are the smallest at the temperatures closest to room temperature (15 °C and 30 °C) and the highest for the temperatures that are most different from room temperature (5 °C and 50 °C). Also, one can notice that deviations at low temperatures are higher for low doses (27 %: 23 %), while at high temperature the measurement error increases for high doses of radiation (25 %: 48 %).

Figure 2 presents the dependence of the measured dose on the temperature when the room temperature calibration is used (full lines), and when calibration at the ampoule temperature is used (dotted line).

CONCLUSIONS

In this paper we examined the temperature influence on the accuracy of measurement of the absorbed radiation dose in the ethanol-chlorobenzene – oscillo-

Table 2. Results of the radiation dose based on the oscillotitrator signal for different temperatures and different calibrations

T = 5 °C	Dose [kGy], room temperature calibration	3.68	3.64	3.64	3.68	3.64	19.38	19.38	19.97	19.18	19.18
	Dose [kGy], calibration on the ampule temperature	5.07	5.00	5.00	5.07	5.00	25.23	26.05	25.23	24.96	24.96
	Expected dose [kGy]	5					25				
	Error [%]	–27.41	–27.33	–27.33	–27.41	–27.33	–23.18	–23.35	–23.18	–23.13	–23.13
T = 10 °C	Dose [kGy], room temperature calibration	4.21	4.21	4.14	4.21	4.14	21.39	22.14	21.59	21.39	21.30
	Dose [(kGy], calibration on the ampule temperature	5.09	5.09	4.99	5.09	4.99	25.56	26.42	25.80	25.56	25.47
	Expected dose [kGy]	5					25				
	Error [%]	–17.29	–17.29	–17.04	–17.29	–17.04	–16.34	–16.20	–16.30	–16.34	–16.36
T = 15 °C	Dose [kGy], room temperature calibration	4.56	4.47	4.51	4.51	4.51	24.17	25.20	24.17	23.73	23.86
	Dose [kGy], calibration on the ampule temperature	5.10	5.00	5.05	5.05	5.05	25.68	26.74	25.68	25.23	25.36
	Expected dose [kGy]	5					25				
	Error [%]	–10.53	–10.64	–10.58	–10.58	–10.58	–5.88	–5.75	–5.88	–5.94	–5.92
T = 20 °C	Dose [kGy], room temperature calibration	5.03	4.99	5.03	5.03	4.99	25.85	25.98	26.12	25.75	25.66
	Dose [kGy], calibration on the ampule temperature	5.03	4.99	5.03	5.03	4.99	25.85	25.98	26.12	25.75	25.66
	Expected dose [kGy]	5					25				
	Error [%]	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
T = 30 °C	Dose [kGy], room temperature calibration	5.15	5.03	5.15	5.15	5.08	29.01	29.36	28.86	28.86	28.72
	Dose [kGy], calibration on the ampule temperature	5.10	4.98	5.10	5.10	5.03	25.77	26.07	25.64	25.64	25.52
	Expected dose [kGy]	5					25				
	Error [%]	1.08	1.06	1.08	1.08	1.07	12.58	12.62	12.55	12.55	12.53
T = 40 °C	Dose [kGy], room temperature calibration	5.88	5.64	5.76	5.76	5.71	32.31	33.17	32.47	32.31	32.21
	Dose [kGy], calibration on the ampule temperature	5.34	5.04	5.19	5.19	5.13	25.57	26.42	25.72	25.57	25.46
	Expected dose [kGy]	5					25				
	Error [%]	10.14	11.78	10.91	10.91	11.25	26.39	25.55	26.23	26.39	26.49
T = 50 °C	Dose [kGy], room temperature calibration	6.55	6.25	6.55	6.50	6.45	37.83	38.31	38.43	37.83	38.91
	Dose [kGy], calibration on the ampule temperature	5.24	5.00	5.24	5.20	5.16	25.51	25.87	25.96	25.51	26.32
	Expected dose [kGy]	5					25				
	Error [%]	25.07	25.12	25.07	25.07	25.07	48.29	48.10	48.05	48.29	47.84

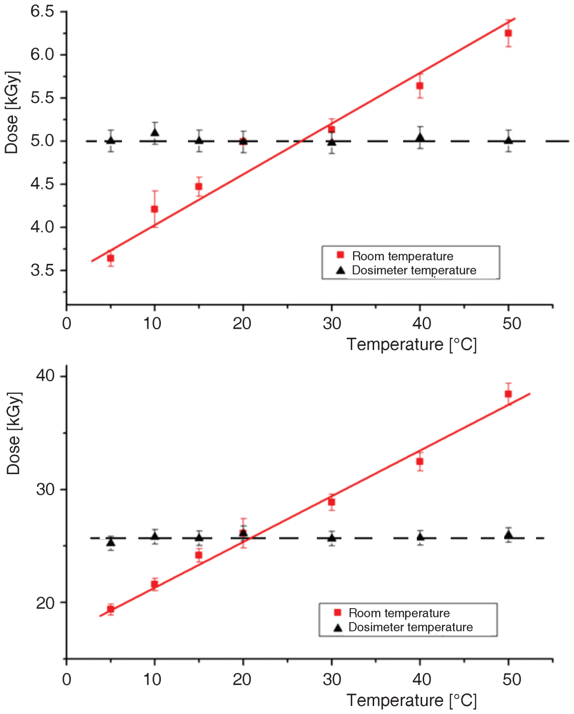

Figure 2. The ratio of calculated dose and temperature for different calibrations

tirtator dosimetry system. We found that the values indicated by the oscillotitrator are highly dependent on the temperature of the dosimeter. The dose calculated from the oscillotitrator signal has to be determined using a calibration curve constructed at the same temperature as the temperature of the ampoule.

The formation of the calibration curve in irradiation facilities' dosimetric laboratories is routinely performed at room temperature, and in this paper, we have shown that the calibration should be performed at the temperature at which the routine measurements are performed later. Routine and reference dosimeters should always be thermostated to the temperature of the calibration curve in order to avoid measurement errors.

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AUTHORS' CONTRIBUTIONS

The manuscript was written by I. T. Vujčić the figures prepared by I. T. Vujčić and S. B. Mašić. Measurement set-up was conceived and prepared by S. B. Mašić and the measurements were performed by I. T. Vujčić. All authors analyzed and discussed the results and reviewed the manuscript. The theoretical analysis was carried out by H. Spasevska, and M. D. Dramićanin.

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ТАЧНОСТ ОДРЕЂИВАЊА АПСОРБОВАНЕ ДОЗЕ ЗРАЧЕЊА КОРИШЋЕЊЕМ СИСТЕМА ЕТАНОЛ-ХЛОРБЕНЗЕН

Осцилотитратор на различитим температурама мерења

Дозиметријски систем етанол-хлорбензен/осцилотитратор често се користи при контроли процеса озрачивања у гама радијационим постројењима. Овај дозиметријски систем је поуздано средство за мерење апсорбоване дозе. Заснован је на процесу радиолитичког формирања хлороводоничне киселине у раствору хлорбензена у етанолу, под утицајем јонизујућег зрачења. Однос температуре озрачивања и оздива дозиметра је комплексна функција која зависи од дозе и температуре за сваку концентрацију хлорбензена. На различитим температурама, покретљивост честица које потичу од хлороводоничне киселине се мења, што доводи до разлике у сигналу на осцилатитратору током високофреквентног кондуктрометријског очитавања. У овом раду смо испитивали утицај температуре на израчунавање дозе апсорбованог зрачења. Показали смо да температура значајно утиче на резултате мерења и да је за добијање прецизних вредности апсорбоване дозе неопходно формирати калибрациону криву на температури озрачивања.

Кључне речи: етанол-хлорбензен, осцилотитратор, дозиметрија, зрачење