

Investigation of high-dose irradiation effects on polystyrene calorimeter response

Farhood Ziaie,
Abbas Noori

Abstract In this work attempts have been made to investigate the variation of polystyrene calorimeter response after frequent irradiations with an electron beam. Polystyrene calorimeters are routinely used in every radiation processing center as a traceable to NPL primary standard dosimeter. Thus, self designed high impact polystyrene as the calorimeter core was irradiated several times up to many thousands of kGy doses. After each irradiation, the specific heat capacity of the polystyrene as the main changeable parameter was measured using the differential scanning calorimeter system (DSC) in the practical temperature range of polystyrene calorimeter. Therefore, correction factors to calculate the precise absorbed dose were obtained. At the final stage, several these calorimeters were irradiated simultaneously along with two Risø standard calorimeters and another correction factor for each tested calorimeter was calculated.

Key words polystyrene calorimeter • electron beam • absorbed dose • high dose • high impact polystyrene (HIPS) • specific heat capacity • differential scanning calorimeter (DSC)

F. Ziaie[✉]
Material and Ion Beam Application Division,
Nuclear Research Center for Agriculture and Medicine,
P. O. Box 31485-498, Karaj, Iran
and Yazd Radiation Processing Center,
P. O. Box 89175-389, Yazd, Iran,
Tel.: +98 261 4411100, Fax: +98 261 4464056,
E-mail: fziaie@yahoo.com

A. Noori
Yazd Radiation Processing Center,
P. O. Box 89175-389, Yazd, Iran

Received: 7 February 2005
Accepted: 15 May 2006

Introduction

High-intensity, high-energy electron beam are widely utilized in the radiation processing of a large variety of industrial products. The standardization of these electron beams is a vital facet of the evaluation of the absorbed dose delivered to the product. Processes or effects on the various materials are achieved only at certain doses, requiring that the dose be well known in order to assure total process completion. Calibration of these electron beams in term of depth-of-cure is a necessary part of an overall quality assurance programme. Various models of calorimeters have been previously constructed in order to address the issues of industrial electron beam accelerator calibrations [5, 8–10]. Calorimeters have been used for making routine and reference dose measurements at electron accelerators for radiation processing [4, 6, 7, 11].

The radiation absorbing polystyrene disk is made of high impact strength grade polystyrene. In this paper, we investigate the variation of polystyrene calorimeter response after frequent electron beam irradiations, up to many thousands of kGy doses.

Experimental

Materials

High impact polystyrene (HIPS) supplied by Tabriz Petrochemical Company with a density of 1.05 g/cm^3 was used in this investigation.

Sample preparation and irradiation

The samples were prepared in the sheet form with $2 \pm 0.1 \text{ mm}$ thickness using the warm press system. The samples in each set were irradiated under the range of doses from 1000 up to 10,000 kGy.

Irradiator

Irradiations are performed with a scanned beam from a high-energy electron accelerator, Rhodotron TT200 type, at a nominal electron energy of 10 MeV, at a beam current of 8 mA, and with a scan width of 100 cm, at a scan frequency of 100 Hz. The accelerator is provided with a variable-speed conveyor to pass the materials under the scanned beam. The distance between the accelerator output windows and the conveyor is about 120 cm.

DSC system

A differential scanning calorimeter system, DSC-50, made by Shimadzu Company, Japan, was used to measure the specific heat capacity of the samples. The DSC system temperature was also calibrated using the standard methods [1]. The temperature range and also the heat flow rate of the system were set up on $0\text{--}100^\circ\text{C}$ and 20 mW, respectively. The reference material used in this work was Al_2O_3 as well.

Gel content

The gel content of the irradiated samples was determined by extracting the sample (m_1) in boiling Tetra Hydro Fouran (THF) for 16 h. After being dried, the insoluble residue (m_2) was weighed. The average gel content was calculated as $100 \times (m_2/m_1)$. Three samples were analyzed to determine the average gel content for a given set of irradiation conditions.

Calorimeter system

The Risø National Laboratory calorimeter system was used to measure the precise dose value to calibrate the tested polystyrene calorimeters. The absorbed dose to the calorimeter core in practice is calculated from the equation given by [2, 5] and [12]:

$$(1) \quad D = (T_2 - T_1 - T_a) \cdot C_{PS} \cdot K_1 \cdot K_2$$

where: D is the absorbed dose in Gy, T_1 and T_2 are the measured temperature before and after irradiation, respectively. The value of the temperatures is determined by extrapolation of the temperature drift rates before and after irradiation to the midpoint of the irradiation period. T_a is a compensative temperature parameter of the conveyor and accelerator structure already measured in our condition to be 0.35°C . K_1 is a correction factor for the change of temperature as a function of time after irradiation where no extrapolation is used. K_2 is a calibration factor derived from inter-comparisons with reference dosimeters. The specific heat capacity (C_{PS}) of the polystyrene is a function of temperature as:

$$(2) \quad C_{PS} = 0.0105 + 1.130 \cdot T \text{ [J/kg}\cdot^\circ\text{C]}$$

For any given calculation involving the specific heat capacity, the average temperature during the irradiation period (i.e., $(T_{\max} + T_{\min})/2$) is used to determine the appropriate value to use in subsequent dose calculation. The beam uniformity for the accelerator was measured by irradiating radio-chromic films with a scanning spectrometer. The electron beam had a good uniformity, being uniform within 2% over the diameter (140 mm) of the calorimeter disk [13]. Before each irradiation, the calorimeter bodies were left to reach a heat equivalent temperature as the irradiated environment. According to the given references, the calorimeter temperature should approximately be constant for about 10 min before irradiation (typically, the change is less than 0.1°C) [1, 9].

Temperature measurement system

The temperature measurement system was a commercial digital read-out unit employing a thermistor sensor. The thermistor was oval in shape, about 1.9 mm in diameter and 4 mm long (model VEC P32A 180, calibrated in Risø).

Measurements

The specific heat capacity measurements of the samples were performed according to the standard [3] using the DSC system. The specimens were sampled by cutting or slicing with a clean knife. Due to the importance of maximizing the contact between the surface of the specimen and specimen holder, it was attempted to make them as small granules as possible. The specific heat capacities of the specimens were calculated with comparing the specimen and the reference material (here is Al_2O_3) base-lines according to the DSC related diagrams (Fig. 1). The specific heat capacity variation of the reference material can be found in the same standard. Therefore, a new correction coefficient (K_3) was calculated which is the ratio of specific heat capacity for a non-irradiated calorimeter core to that irradiated at high doses.

At the final stage, several self designed calorimeters were irradiated simultaneously along with two Risø

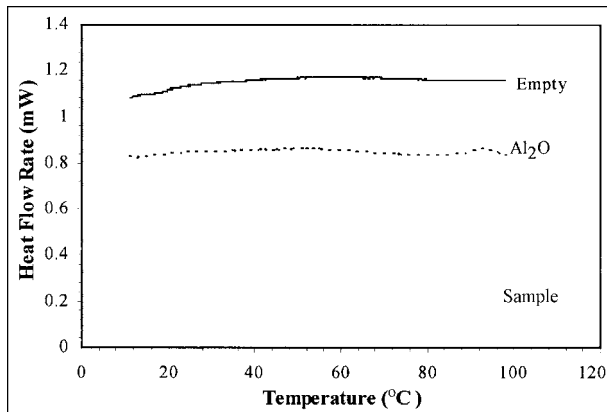


Fig. 1. DSC diagram showing the base-line of the sample, reference material and the empty cell (specimen holder).

standard calorimeters and the correction factor (K_2) for each tested calorimeter was calculated. To get a reliable result, the irradiations were made at four different doses of 8, 20, 30, and 40 kGy.

Results and discussion

Figure 2 shows the K_3 coefficient variation of the HIPS against the different irradiated doses for the same temperature. The gel content of the irradiated samples was measured to see the amount of cross linking induced by the radiation (Fig. 3). In fact, the cross linking process

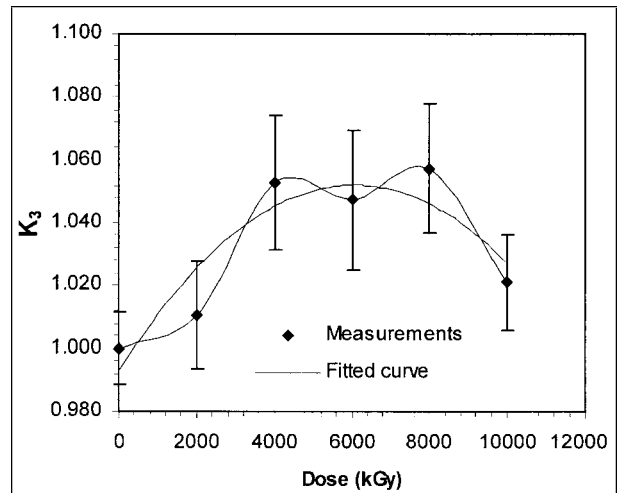


Fig. 2. Variation of K_3 coefficient of polystyrene calorimeter vs. different irradiated doses.

means the forming of bigger molecules or increasing the molecular weight of the polymers. The result shows that the HIPS were cross-linked up to about 4–5 MGy radiation dose without degradation. It also means that the cross linking process is saturated in the polymer media. Thus, at a dose higher than the mentioned value the chain scission will be the dominant process and so it will cause decreasing of molecular weight, consequently. Therefore, the specific heat capacity should change proportionally to the molecular weight because

Table 1. Correction factors (K_2) for eleven self designed calorimeters and the overall average correction factor

No. of tested calorimeter	1	2	3	4	5	6	7	8	9	10	11					
No. of experiments	30.6	30.2	28.8	29.4	31.1	28.2	8.3	9.8	9.2	9.0	20.4	21.4	20.4	40.8	40.6	41.0
Risø standard measured dose [kGy]	0.943	0.948	*	0.973	0.944	0.928	0.936	0.919	0.956	0.920	0.952	*	0.941	0.939	*	0.919
	0.890	0.960	0.939	0.968	0.957	0.923	0.915	0.927	0.962	0.920	0.933	0.952	0.944	0.946	0.938	0.922
	0.919	0.950	0.898	0.967	0.934	0.940	0.907	0.933	0.964	0.924	0.930	0.934	0.931	0.926	0.929	0.940
	0.917	0.938	0.904	0.957	0.953	0.934	0.931	0.930	0.964	0.928	0.933	0.948	0.945	0.941	0.920	0.928
	0.891	0.965	0.902	0.964	*	0.936	0.939	0.930	0.956	0.927	0.933	0.934	0.940	0.929	0.920	0.922
	0.922	0.941	0.927	0.963	0.977	0.956	0.921	0.922	0.935	0.922	0.922	0.939	0.927	0.929	0.928	0.930
	0.915	0.943	0.996	0.953	0.955	0.935	0.909	0.936	0.935	0.909	0.933	0.926	0.936	0.940	0.927	0.928
	–	0.949	0.903	0.946	0.952	0.935	0.905	0.916	0.935	0.907	0.922	0.932	0.929	0.919	0.919	0.914
	0.935	0.940	0.899	0.947	0.946	0.919	0.934	0.909	0.932	0.901	0.938	0.924	0.928	0.917	0.953	0.908
	1.028	0.942	0.944	0.960	0.954	0.932	0.915	0.934	0.956	0.926	0.932	0.942	0.906	0.934	0.954	0.913
	0.895	0.946	*	0.963	0.931	0.920	0.921	0.926	0.944	0.922	0.940	0.936	0.953	0.906	0.921	0.925

$$\bar{K}_2 = 0.933, \sigma = \pm 0.019 \equiv 2\%.$$

* Due to some problems during the measurement, the resulted value was omitted.

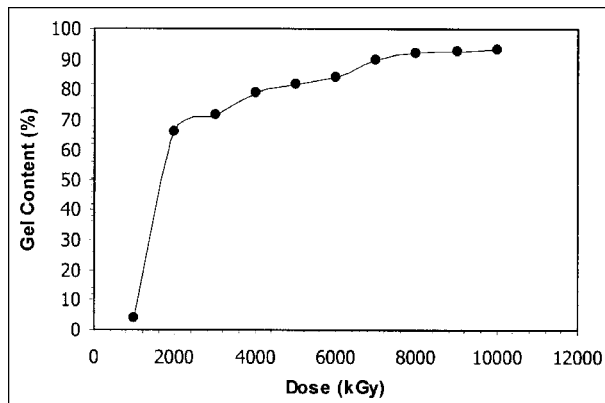


Fig. 3. Gel content of the irradiated samples.

of the fact that the same incident energy can increase the temperature of the medium with higher molecular weight, less than lower molecular weight, due to the less chain/molecule mobility/vibration and the high chain/molecule mobility/vibration, respectively. It means that the molecular weight increment cause the heat capacity to increase, and *vice versa*. Therefore, the K_3 coefficient will vary, accordingly. The calibration results of eleven self designed calorimeters against the Risø standard calorimeters are indicated in Table 1, as well.

Conclusion

The result show that the calorimeters measuring the dose values will change by a few percent after absorbing some thousands of kGy doses, due to the specific heat capacity variations of HIPS. Therefore, to have an accurate absorbed dose measurement by a polystyrene calorimeter a correction factor must be involved periodically depending on the routine use. The final equation to calculate the absorbed dose in the polystyrene calorimeter can be presented by:

$$(3) \quad D = 0.933 \cdot (T_2 - T_1 - T_a) \cdot C_{PS} \cdot K_3$$

where: C_{PS} is the specific heat capacity of non-irradiated polystyrene calorimeter core. Also because of T_1 and

T_2 are the extrapolated temperature thus, it is assumed that $K_2 = 1$ in Eq. (1).

References

1. ASTM (1992) Standard practice temperature calibration of differential scanning calorimeters and differential thermal analyzers. ASTM E 967
2. ASTM (1995) Standard practice for use of calorimetric dosimetry systems for electron beam dose measurements and dosimeter calibrations. ASTM E 1631
3. ASTM (1995) Standard test method for determining specific heat capacity by differential scanning calorimetry. ASTM E 1269
4. Fielden EM, Holm NW (1970) Dosimetry in accelerator research and processing. In: Holm NW, Berry RJ (eds) Manual of radiation dosimetry. Marcel Dekker, New York, pp 297–300
5. Humphreys JC, McLaughlin WL (1990) Calorimetry of electron beams and the calibration of dosimeters at high doses. Radiat Phys Chem 35:744–749
6. Icre P (1983) Industrial dosimetric controls of ionizing treatments by electron beam. J Indust Irradiation Tech 1:163–178
7. Janovsky I (1985) Dosimetry methods applied to irradiation with Tesla-4 MeV linear electron accelerators. In: High-dose dosimetry. IAEA, Vienna, pp 307–316
8. McLaughlin WL, Boyd AW, Chadwick KH, McDonald JC, Miller A (1989) Dosimetry for radiation processing. Taylor and Francis, London
9. Miller A, Kovacs A (1985) Calorimetry at industrial electron accelerators. Nucl Instrum Methods Phys Res B 10/11:994–997
10. Miller A, Kovacs A (1990) Application of calorimeters for routine and reference dosimetry at 4–10 MeV industrial electron accelerators. Radiat Phys Chem 33:774–778
11. Panta PP, Bulhak Z (1985) Statistical and metrological aspects of 20 years experience of radiation processing in Poland. In: High-dose dosimetry. IAEA, Vienna, pp 47–60
12. Risø National Laboratory (1996) Absorbed dose measurement with graphite calorimeters. In: Quality manual, QC 2422-1.4, 4th ed. Risø National Laboratory, Denmark, pp 1–4
13. Ziaie F, Afarideh H, Hadji-Saeid SM, Durrani SA (2001) Investigation of beam uniformity in industrial electron accelerator. Radiat Meas 34:609–613