Challenges of Using Radiochromic Films as Dosimeters for Low Voltage Electron Beams

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Abstract

Thin radiochromic films are commonly used to determine the quantity of electron dose applied to surfaces. The advantages of these films are that they are widely available, inexpensive and require relatively simple equipment to process and measure. However, there are many challenges when users attempt to use them as accurate and repeatable metrology tools for low voltage (≤ 150kV) electron beams (LVEB). In this paper we report on the results of a study of the stability, precision, repeatability and accuracy of using two common brands of radiochromic films and make recommendations for exposure, curing and measurement procedures to allow the most accurate and precise measurements to be made of low voltage electron beam dose.

Introduction

Low voltage electron beams (LVEB) with acceleration ≤ 150kV are being increasingly used in manufacturing applications including: curing thin film (typically <25μm) inks and coatings; reducing pathogens on food and beverage containers; sterilizing surfaces of medical devices; modifying chemical and mechanical properties of polymer surfaces; and treating air for sterilization and for organic pollution abatement.   With this transition into manufacturing environments come more stringent requirements for the quality of dose measurement. Electron beam equipment manufacturers characterize and quantify the dose rate of their products during their manufacturing process. Process manufacturers employ statistical process control (SPC) methods that require stable metrological tools to indicate process shifts and determine whether a products stay within specification. Typical manufacturing processes in LVEB applications are designed for maximum process variation of ±10%. Proposed requirements for dose measurement systems to support this target are shown in Table I.

Table I: Dose Metrology System Requirement

Category	**Requirement**	**Comment**
Precision & Repeatability	≤ 1%, 1·σ	Enable Manufacturing SPC methods to monitor production stability
Accuracy	≤ 3%, 1·σ	Critical for aseptic processing
Spatial Resolution	≤ 2mm	Enable dose mapping 3-d objects
Range & Sensitivity	1 – 1000kGy ≥ 1:1 response < 10μm	Cover all low & high dose applications Indicate small process changes Measure surface dose
Temporal & Environment Stability	Read Time: 14 days T: 10-40°C, RH: 20-70%	Remote sites need to ship dosimeters before reading Industrial manufacturing conditions

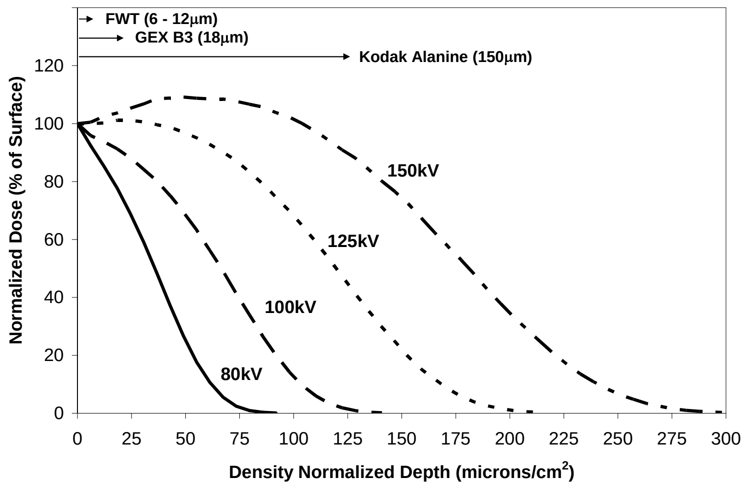


Figure 1: calculated dose-depth curves for LVEB irradiating unit density material.

LVEB delivers dose to the surface of materials. Depth-dose curves are provided in Figure 1 showing electrons irradiating unit-density material. Note that the distribution of dose into the material strongly depends upon the accelerating voltage: 80kV beam has a steep gradient in the near surface (top 10µm) region; 150kV beams produce flatter profiles out to 100µm. To measure dose at the surface of the process with minimal impact due to this dose gradient requires a sensor of high depth resolution.

The most common dose measurement systems employ films less than 25µm thick and doped with radiochromic dye. The dye causes the film to change color upon exposure to energetic radiation. The intensity of the color of the film, measured as by its optical density at a fixed wavelength, indicates the amount of absorbed dose. The optical density is commonly normalized to the film thickness by ratioing OD to T (OD/T) in order to compensate for the related effects; for example, a thicker film would absorb more beam energy than a thinner film as more material is in the path of the radiation. The typical measurement protocol consists of the following steps:

1. pre-exposure optical density measurement (OD_i)
2. irradiation of the dosimeter
3. curing of the dosimeter to ensure complete optical density change and stabilization
4. post-exposure optical density measurement (OD_f)
5. film thickness measurement (T)
6. calculation of primary absorbed dose metric: $(OD_i - OD_i)/T = \Delta OD/T$
7. conversion of metric to absorbed dose using a calibration equation or curve

The purpose of this work was to study the precision, repeatability and accuracy of common radiochromic films and to make recommendations for exposure, curing and measurement procedures to allow the most accurate and precise measurements to be made of low voltage electron beam dose.

Experimental Methods

Strategy

Since this work was undertaken to understand the practical capabilities of the most common LVEB dosimetry systems, it was essential to minimize or eliminate the error from outside factors, especially from the dose exposure system. All dosimeters were exposed to gamma radiation from a well-characterized ^{60}Co source. We selected gamma radiation over methods such as high voltage electron beam and x-radiation for the following reasons: 1) gamma provides highly-controlled and traceable, simultaneous dose to the dosimeter samples; 2) it is the benchmark for radiation-based terminal sterilization; 3) it provides uniform dosing of the entire thickness of the dosimeter film; and 4) it is available from many national laboratories, including the National Institute of Standards and Technology (NIST)[1]. The primary drawbacks of using gamma radiation are its expense and the difference in dosing rate compared to electron beam irradiation. The former is not a technical consideration but does limit the use of gamma for primary LVEB dosimeter calibration. The latter effect may be important; however, the benefit of traceability of gamma radiation was deemed more important for this work.

Equipment & Methodology

Dosimeters

Dosimetry films from two suppliers were studied in this work.

1. Far West Technology[2], part FWT-60-810, lot 1088 (FWT). This dosimeter employs a hexa(hydroxyethyl) aminotriphenylacetonitrile (HHEVC) dye cast in nylon 6/6 polymer base. The film changes during irradiation from clear to deep blue; change in optical density is measured at either 510 or 600nm. Nominal thickness of the films is specified as 8 – 12μm.
2. GEX Corporation[3], part B3103, lot BB (GEX). This dosimeter employs the B3 film stack based on pararosaniline cyanide and developed at Riso National Laboratory[4]. The film changes during irradiation from clear to deep pink; change in optical density is measured at 554nm. Nominal thickness of the films is specified as 17 – 20μm.

Films were purchased as bare sheets and were stored in amber bags until used. The lighting in all laboratory areas used for dosimeter preparation and measurement were filtered to prevent light of wavelength <400nm from reaching work surfaces.

It should be noted that a thin film alanine dosimeter is also available. It was not selected for this work due to the lack of ready access to the required measurement equipment.

Five hundred individual dosimeter tags were prepared from each type of film after gamma irradiation. A paper frame was attached to the dosimetry film to provide mechanical support; a 0.25 inch (6mm) hole was present to permit measurement of optical density change.

Dose Exposure

Stacks of dosimetry sheets, enough to produce at least 40 tags at each dose and 500 total, were sent to NIST for irradiation in amber bags to minimize unintended irradiation. Each stack of film received one of the following total absorbed gamma doses: 2, 5, 10, 20, 30, 50, 75, 100, 125, 160 or 200kGy. Irradiated dosimeters were returned by NIST in amber bags using rapid delivery service. Control sample tags were also sent in the same package to NIST but did not receive gamma exposure. This allowed us to compensate for inadvertent irradiation of the dosimeter films due to security screens and environmental effects.

Stabilization (Curing)

Dosimeter tags were stabilized at AEB by curing at room or elevated temperature for varying amounts of time. Elevated temperature curing was performed in a Thermolyne Type 37900 culture incubator. Room temperature and humidity during sample preparation and analysis were recorded but not controlled.

Optical Density Measurement

Manufacturer's instructions were followed for each type of dosimeter film. FWT dosimeters were measured at 510 and 600nm using the FWT-92D photometer calibrated using filters of known optical density. GEX dosimeters were measured at 554nm using a Spectronic/Unicam Genesys 20

Spectrophotometer. The optical density of the control tags was measured at AEB and used as the OD_i value. No statistically significant change in OD_i was observed before and after shipment to and from NIST. All gamma irradiated dosimeter tags were measured after irradiation and stabilization.

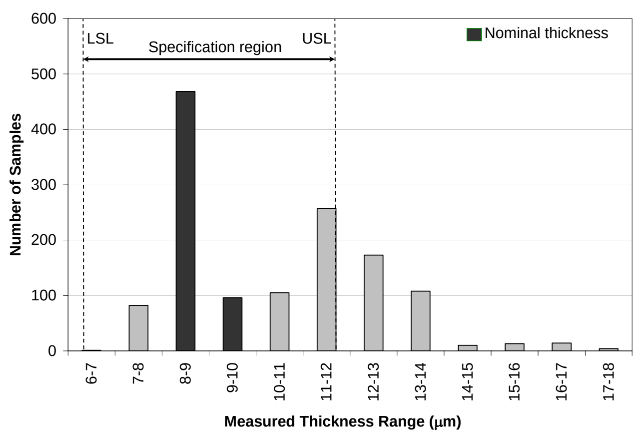
Dosimeter Thickness Measurement

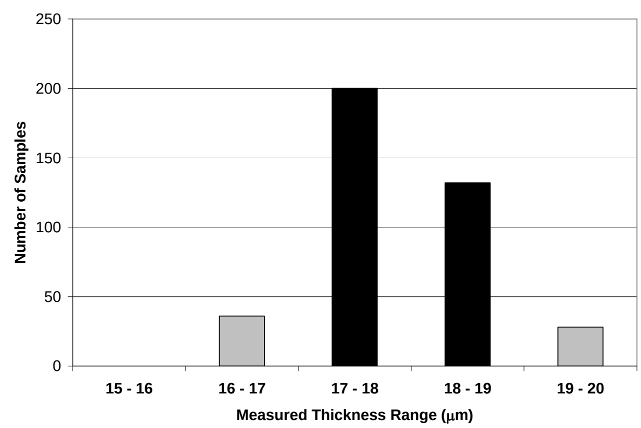
The thickness of each dosimeter was measured by optical reflectance using a Filmetrics F20 Optical Measurement System. A detailed evaluation of optical reflectance vs. contact profilometry has been performed and will be reported in a forthcoming paper. Total repeatability in thickness using this system is <0.1% ($1 \cdot \sigma$).

Results

Dosimeter Thickness Variation and Its Effects

In order to study the effect of dosimeter thickness on measured absorbed dose, the distribution of thickness was measured for each type of film. Figures 2 and 3 show the results for the FWT and GEX dosimeter tags prepared for this work. Note that the FWT dosimeters show a much wider range of thickness than the GEX. Also, a significant population of the FWT tags (25%) was outside of manufacturer's specification; some of the tags in this population were nearly 100% thicker than the target value.

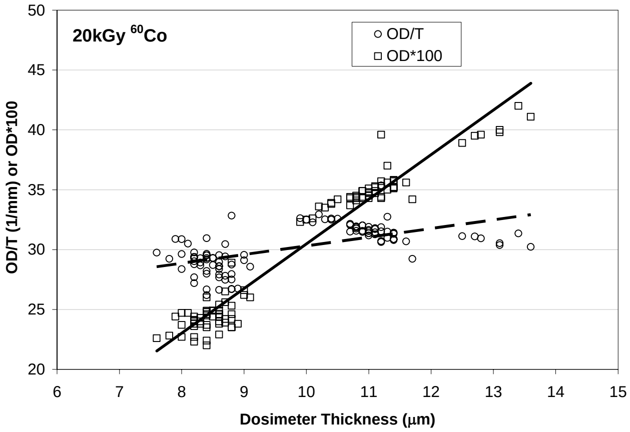


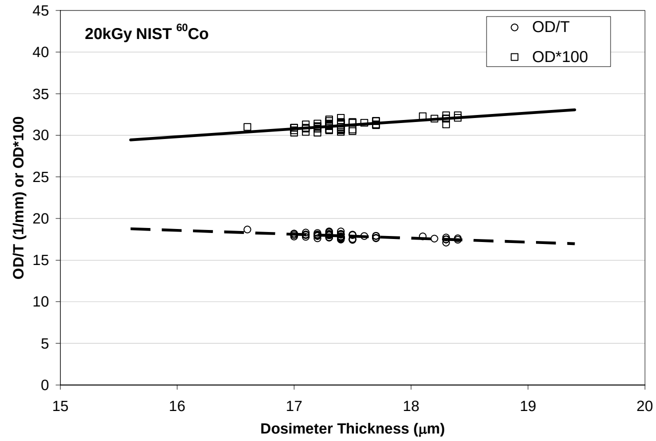


Figures 2 and 3. Distribution of thicknesses of FWT dosimeters (left) and GEX dosimeters (right).

Optical Density Change

As mentioned earlier, optical density is often normalized by dosimeter thickness in order to compensate for any related affects on measurements. If this normalization is effective, the absorbed dose metric, OD/T, should be independent of dosimeter thickness. Figures 4 and 5 shows the results of testing the OD/T normalization for FWT and GEX dosimeters exposed at 20kGy, respectively. The squares show the optical density data without thickness correction (OD*100); the circles show the effect of normalization. As expected, optical density shows dependence on thickness for both types of dosimeters; the effect is stronger for FWT than GEX. However, in both cases there is still a noticeable dependence of OD/T on dosimeter thickness: the FWT dosimeters are under-compensated while the GEX dosimeters are over-compensated. In both cases, the thickness of the dosimeter affects the OD/T result indicating that the final dose calculated will not be independent of the thickness. Therefore, both dosimeters' thicknesses would cause systematic errors in dose measurement, even when calibration curves using OD/T are employed. The effect is lesser for the GEX system.





Figures 4 and 5. Dependence of optical density change (OD) and normalized metric (OD/T) on thickness for 20kGy exposed FWT (left) and GEX (right) dosimeters.

Precision & Repeatability – Metric Distribution

Since all dosimeter tags in a given batch were simultaneously irradiated with ^{60}Co, all tags should have received the same thickness-normalized absorbed dose and therefore demonstrated the same metric within the error of the optical density and thickness measurements equipment. In a forthcoming paper[5] we will show that the combined machine random standard error for these measurements is less than 0.2%. The distribution of the measured data from Figure 4, FWT dosimeters irradiated to 20kGy absorbed dose, is shown in Table 2. Note that the 1·σ standard error for the combined data using all dosimeter thickness is nearly 6%. However, the standard error can be significantly reduced if dosimeters are binned by thickness. Clearly dosimeters of similar thickness must be used for the best possible precision in thin film dosimetry measurements.

Table 2: Distribution of thickness-normalized dose metric, OD/T

Bin Size (µm)	Standard Deviation, 1·σ	Standard Error
All bins	1.8	5.9%
7 – 8	1.0	3.3%
8 – 9	1.3	4.5%
10 – 11	0.5	2.6%
11 – 12	1.1	3.6%
12 – 13	0.1	0.3%
13 – 14	0.5	1.6%

Stabilization Protocol Effects

In order to drive to completion the chemical changes caused by radiation absorption, stabilization (often called curing) is recommended by dosimeter film manufacturers. The effect of stabilization conditions on the absorbed dose metric was studied in detail for the FWT dosimeters. Subsets of dosimeter tags given the same absorbed dose were subjected to one of the following stabilization processes as recommended by the manufacturer:

- Room temperature exposure for 24 hours
- 65°C exposure for 5 minutes
- 65°C exposure for 15 minutes

- 90°C exposure for 2 minutes

Figure 6 shows the effect of stabilization on OD/T; all dosimeters were read immediately after returning to room temperature. Note that the stabilization protocol has a strong effect on the measured OD/T and that the effect becomes exaggerated at higher absorbed dose. The faster, higher temperature stabilization (90°C, 2 minutes) results in the largest OD/T; stabilization at 65°C for 15 minutes had the lowest OD/T. This may be an indicator of the kinetics of the chemical reaction that induces the color change in the film. Clearly one must select a single stabilization protocol and use it exclusively in order to prevent systematic error in absorbed dose measurements.

The importance of a complete dosimetry measurement protocol becomes even clearer when reviewing the results of studying the impact post-stabilization delay time on OD/T. Figure 7 documents the change in OD/T when dosimeters are measured immediately after stabilization (as in Figure 6) and after waiting 1, 2 or 24 hours. The absorbed dose metric changes by up to 4% in 24 hours. The delay between stabilizing and reading dosimeters must be carefully controlled in order to avoid additional dose measurement errors. This effect also has implications for the accuracy of measurements that are made by exposing the dosimeters in one location and shipping, stabilizing and reading the dosimeters in a second location. Care must be taken to standardize all time delays to match with a calibration curve developed under an identical protocol.

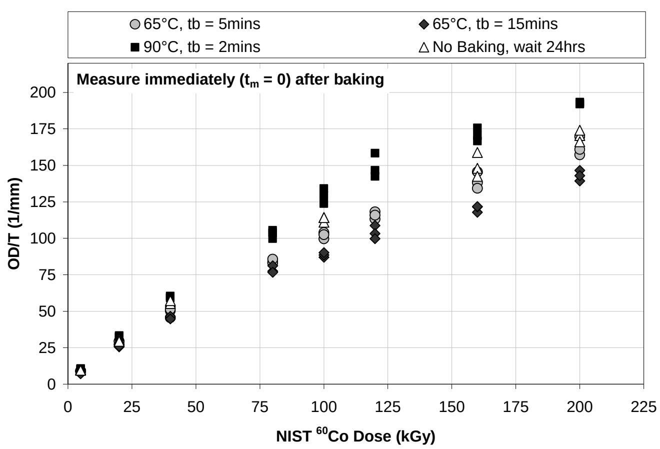


Figure 6. Impact of stabilization protocol on measured OD/T.

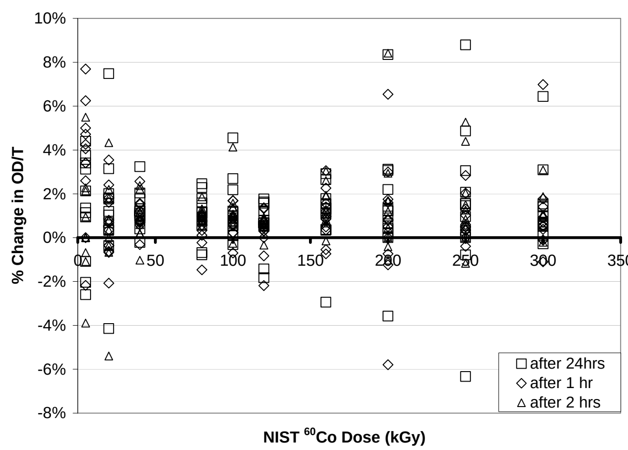


Figure 7. Impact of post-stabilization measurement time.

Dose Sensitivity

Figure 8 shows the response of the OD/T metric to absorbed dose. Note that as expected the response is non-linear; at higher absorbed doses the dye chemistry is saturated. FWT is reported[3] to saturate for doses >200kGy while GEX saturates[4] at absorbed doses > 100kGy; these data are consistent with these reports. Note the increased distribution in OD/T at higher dose; this is partially due to the distribution of dosimeter thicknesses but is not completely explained by that effect. Figure 9 expands the graph to emphasize the lower dose region of 10 to 30kGy. Many of the manufacturing applications that employ LVEB operate in this regime. Note for this narrow regime the FWT dosimeter sensitivity is greater than 1 while GEX is approximately 0.75. The GEX data has tighter distribution at each dose point. GEX would be preferred dosimetry system for higher precision measurements due to the repeatability of OD/T metric and the narrower distribution of dosimeter thicknesses. FWT has the advantage of slightly higher sensitivity and has a broader dose sensitivity range.

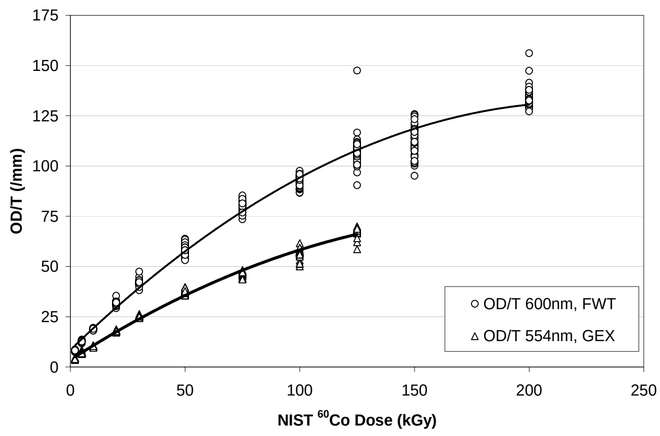


Figure 8. Sensitivity of OD/T to absorbed dose – full range.

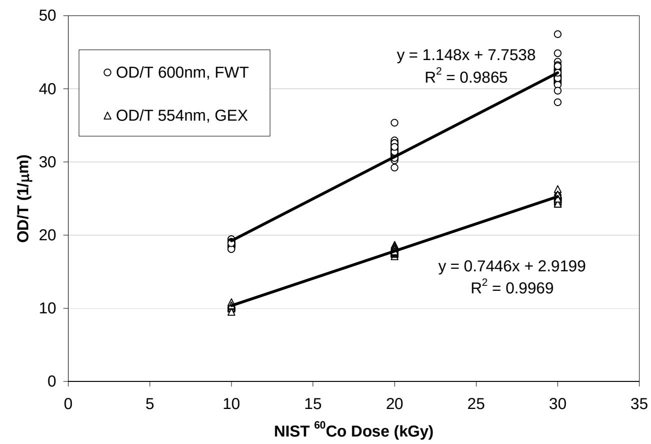


Figure 9. Sensitivity at lower dose regime.

Calibration curves have been developed for both types of dosimeters and are in active use in our electron beam system manufacturing and applications development. When high precision is required, dosimeters are binned to within 1µm for each measurement; stabilization is performed at 65°C for 5 minutes; and measurement is performed within 1 hour of stabilization.

Summary and Implications

A systematic study of the equipment and methods that will impact the stability, precision, repeatability and accuracy of absorbed dose measurements made using two common brands of radiachromic films has been completed. Gamma radiation was used as the standard irradiation source due to its traceability and control. Dosimeters made from FWT film had a wide distribution of thicknesses; the impact of the thickness on the absorbed dose metric could not be adequately corrected by simple normalization. Dosimeters made from GEX film also showed this effect but to a much smaller degree. Binning of dosimeters into batches of similar thickness for a given measurement is important. Post-irradiation stabilization protocol and measurement delay time have a strong impact on the absorbed dose metric and must be standardized and controlled in order to give the most precise and accurate dose measurements. The two types of dosimetry films have complementary strengths: one is more sensitive in the low dose (<30kGy) regime and has a wider dose response range; the other has better overall precision. Clearly neither system meets all of the requirements for a stable, accurate and precise metrology system which would enhance confidence in delivered dose as the adoption of low voltage electron beam systems accelerates in manufacturing applications.

Acknowledgments

The authors thank J. Puhl of the National Institute for Standards and Technology (NIST) for irradiating the samples under for-fee contract. The assistance of L. Chingozha, a summer student from the Chemical Engineering Department of the Massachusetts Institute of Technology (MIT), was essential to this work. Chingozha was resident in our facility as participants in the 2007 MIT Undergraduate Practice Opportunities Program.

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