Characterization and real-time optical measurements of the ionizing radiation dose response for a new radiochromic medium

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A new radiochromic film, GafChromic EBT, was investigated for use in a real-time radiation dosimetry system. It was found to be approximately eight times more sensitive to ionizing radiation dose, exhibited less postexposure development and achieved stable readout faster than one of its predecessors, GafChromic MD-55. A clear distinction in change in optical density between exposure and postexposure was observed, but the measurements obtained during exposure were not linear with time or dose. This could not be explained by a shift in wavelength of maximum change in absorbance, as it was stable at ~636 nm during the entire exposure range (up to 9.52 Gy). Increasing the spectral window of interest over which calculations were performed did little to correct the nonlinearity. The radiochromic film exhibited small dose rate dependence in real-time measurements, with an increase in standard deviation of change in optical density measurements from 0.9% to 1.8% over a sixfold variation in dose rate. Overall, GafChromic EBT has increased sensitivity and decreased postexposure darkening, and this bodes well for its potential role as a radiation dosimeter, including real-time applications. © *2005 American Association of Physicists in Medicine*. [DOI: 10.1118/1.1951447]

I. INTRODUCTION

Several radiochromic films, manufactured by International Specialty Products (ISP, Wayne, NJ) for dosimetry purposes, have been studied for more than a decade,[1–3] often for two-dimensional (2D) radiation dose verification,[4,5] and thorough reviews have been published.[6,7] In general, a dose of 1 Gy or more is recommended for accurate dose measurement,[8] preventing use of these films for low-dose studies. Radiochromic substances have also recently been investigated in real-time dosimetry systems.[7] In these studies, GafChromic® MD-55 performed reasonably well whereas some issues remained unresolved. GafChromic® EBT film (ISP) was created for use in external beam dose verification, and is advertised by the manufacturer to be more sensitive than both GafChromic® HS and MD-55 films (referred to as EBT and MD-55 from here on, respectively). This makes it potentially useful for low dose verification, such as doses delivered in each intensity-modulated radiation segment. It is also proposed as an improvement for some of the real-time dosimetry issues, including stability of wavelength of maximum absorbance (λ_{max}),[9] and decreased extent of postexposure darkening.

The radiochromic material used in EBT film appears to be a reasonable candidate for use as an optical media for real-time *in vivo* dosimetry, as per criteria previously listed.[7] It addresses the requirement for a small radiation sensitive volume with improved water-equivalency compared to its predecessor MD-55, increased sensitivity to low doses, and faster polymerization kinetics, resulting in a stable response shortly after exposure (ISP product information). To determine the suitability of EBT to real-time *in vivo* dosimetry and verify some of the above-mentioned claims, an experimental setup and method previously used in real-time investigations of MD-55 film was employed.[7] In this technical report investigations of linearity, stability, and sensitivity are described.

II. METHODS AND MATERIALS

The experimental setup described previously[7] was modified by removing the beam splitter [Fig. 1(a)] and using a light emitting diode (HLMP-ED25-TW00, Agilent Technologies, Palo Alto, CA) with a ~633 nm emission peak [Fig. 1(b)], chosen to interrogate the greater of the two absorbance peaks of the EBT film. The EBT film used in these investigations (Lot No. 34098-8×2W) consisted of five layers (Fig. 2, ISP product information) where the clear polyester was assumed to be Mylar™ as employed in the MD-55 film.[2]

The system operated with 13.55±0.01 mA power supply driving light output from the light emitting diode. A dark spectrum (I_D) and a reference spectrum (I_R) were collected prior to each radiation exposure. The I_R at a given wavelength is proportional to the radiant power of the light transmitted through the unirradiated piece of EBT film (measured over ~0.33 mm^2).[7] Irradiation of the film was initiated shortly (typically <3 s) after starting the collection of the sample spectra (I_s). The spectrometer integration time ranged from 8 to 10 ms, depending on the study performed, and each spectrum was recorded. The change in absorbance (ΔA), at any measurement interval, for each wavelength was calculated using

$$\Delta A(\lambda) \equiv \log_{10}\left(\frac{I_R(\lambda) - I_D(\lambda)}{I_S(\lambda) - I_D(\lambda)}\right). \quad (1)$$

The EBT film likely undergoes interactions with interrogating light other than absorption. However, the optical scatter-

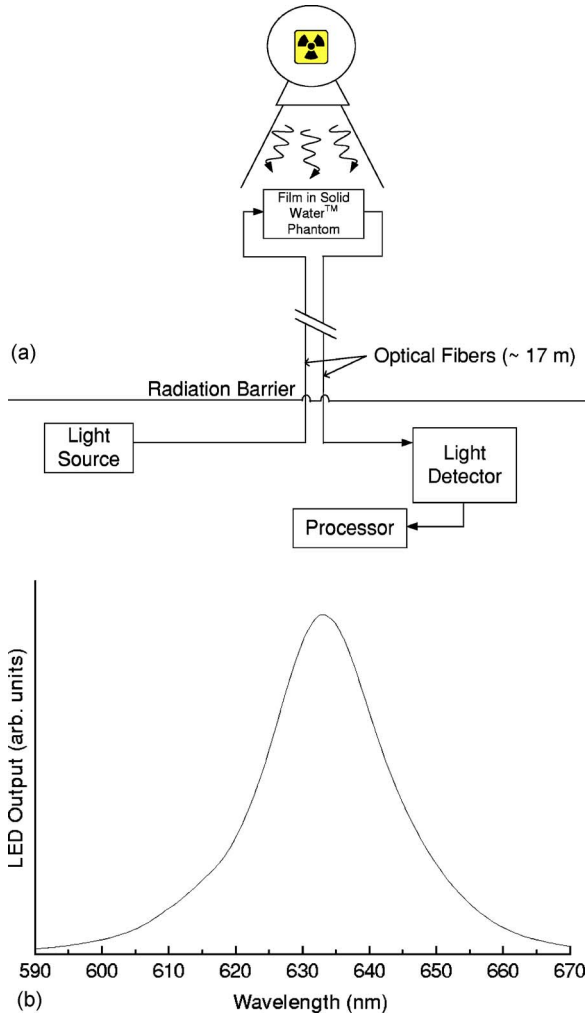

FIG. 1. (a) Schematic of experimental setup. Detector is an Ocean Optics Inc. SD2000 dual-channel spectrophotometer. Processor is a computer with Ocean Optics Inc. IOBase32 and Matlab® 6.1. (b) Measured emission of the light emitting diode used in experimental setup.

ing was assumed to be negligible, and the fraction of light reflected from both the clear polyester layer and from the sensitive layer was assumed to be independent of dose. Hence, the change in absorbance as measured by light transmission is assumed to be entirely due to radiation induced increase in concentration of absorbers within the sensitive layers of the film.

97 μm	clear polyester
17 μm	radiation sensitive layer
6 μm	deposition layer
17 μm	radiation sensitive layer
97 μm	clear polyester

FIG. 2. Schematic of layers in EBT film. The overall atomic composition of this configuration is: 42.3% C, 39.7% H, 16.2% O, 1.1% N, 0.3% Li, and 0.3% Cl (ISP product information).

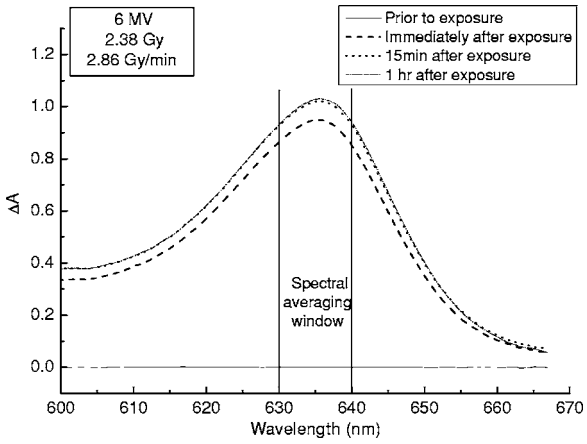

FIG. 3. Change in absorbance of EBT film over a range of wavelengths before, immediately after, and at two time points postexposure.

The method is illustrated in Fig. 3, which shows the change in absorbance of a single piece of EBT film prior to exposure, immediately after exposure to 2.38 Gy at 2.86 Gy/min, and at 15 and 60 min after the completion of exposure. The spectral "window" of interest, or range for optical density calculation, is a 10 nm band around the main peak (630–640 nm). The change in optical density (ΔOD) was then defined as

$$\Delta \mathrm{OD} \equiv \frac{1}{\lambda_n - \lambda_1} \times \sum_{i=1}^{n-1} \left(\frac{\Delta A_i + \Delta A_{i+1}}{2} \right) (\lambda_{i+1} - \lambda_i), \qquad (2)$$

where λ_1 to λ_n are wavelengths that span the window of interest in the spectrum, sampled three times per nanometer.

The light power incident onto a ~650 μm diameter spot[7] of the film within the holder was measured (840-C power meter and 818-SL detector, Newport, Mountain View, CA) to be ~75±10 nW. The effect of this power level of interrogation light on GafChromic® EBT film was investigated by monitoring the ΔOD for an unexposed piece of film over a period of one hour on eight separate occasions. The average increase in OD was 0.001±0.002, and considered statistically insignificant (type I error of 5%, where type I error would occur if a hypothesis where the increase in OD is null is rejected, even though it was true).[10]

The radiation exposures were performed using the same setup as described in real-time investigations of MD-55.[7]

A. ΔOD of EBT film versus time

Five 1 cm×1 cm pieces of EBT film were exposed to 9.52 Gy at an average dose rate of 2.86 Gy/min. The transmitted spectra were collected during exposure, and for approximately one hour after completion of exposure. The ΔOD values were calculated as described previously and plotted versus time to investigate suitability of obtained signal with respect to a *Fast Kinetics* model.[7]

B. Sensitivity and stability comparison between EBT and MD-55 films

One 1 cm×1 cm piece of EBT and four stacked 1 cm ×1 cm pieces (to increase optical signal and minimize error due to small fluctuations in light signal)[7,11] of MD-55 (Lot No. L1906MD55) were each exposed to a dose of 1.9 Gy. The MD-55 film was optically interrogated with a 680 nm light emitting diode (Roithner Lasertechnik, Vienna, Austria), using the same setup as illustrated in Fig. 1(a), and the same spectral range as in previous investigations (670–680 nm).[7] The 33.0±0.3 nW of power delivered to a ~650 μm spot on the previously unirradiated film was shown to have a small effect on ΔOD (0.0034 over 24 hours), but was tolerated in order to keep the signal intensity and integration time on the spectrophotometer sufficient for real-time measurements. For both types of radiochromic film, spectra were obtained during exposure and for approximately 19 h following exposure without disturbing the system. The data obtained for MD-55 film were divided by 4 (corresponding to the factor of 4 increase in sensitivity anticipated for the four layers used in this study) in order to obtain the average ΔOD increase for each individual film. To compare stability between the two types of films, the postexposure measurements were normalized by the change in optical density measured immediately at the end of exposure.

C. Dependence of real-time ΔOD measurements on dose rate for the EBT film

Dose rate dependence of ΔOD measurements performed during, or immediately at the end of, radiation exposure was investigated by irradiating EBT film to the same dose of 9.52 Gy at one of two different dose-rates (5.71 or 0.95 Gy/min). Five films in total were exposed at each dose rate. The ΔOD as a function of time was converted to a function of dose during exposure, using average dose rates as listed above. The values recorded immediately at the end of exposure were compared (analysis of variance using type I error α=0.01).[10]

D. Structure of active crystals in MD-55 and EBT films

The active layer suspensions from MD-55 and EBT films[9] were imaged at the Advanced Optical Microscopy Facility (Ontario Cancer Institute, Toronto, Canada). A differential interference contrast and Plan-Apo 63x/1.4 NA lens on an inverted microscope (Axiovert 200M, Carl Zeiss, Oberkochen, Germany) were used, providing 0.22 μm resolution.

III. RESULTS AND DISCUSSION

A. ΔOD of EBT film versus time

If radiochromic substances are to be used in real-time dosimetry, errors due to postexposure darkening have to be accounted for. The simplest way to eliminate a substantial fraction of postexposure darkening is to limit measurements to those performed during exposure. This requires knowing when the radiation is present. Figure 4 shows ΔOD versus time during exposure to 9.52 Gy (at 2.86 Gy/min) and for approximately one hour after exposure for a single piece of EBT film. A distinct difference in darkening rates between exposure and postexposure (intra- and interexposure, respectively) can be seen, and duration of radiation exposure can be easily deduced from the signal.

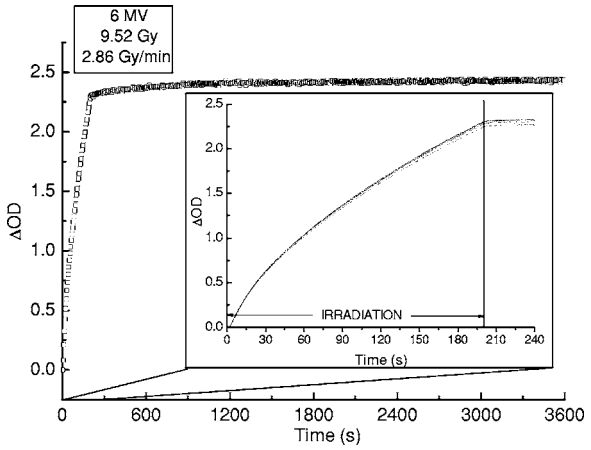

FIG. 4. Change in OD vs. time for a single piece of EBT film (beam on at 0 s, off at ~200 s); change in OD vs. time for five pieces of EBT film shown on a reduced time scale (inset).

Change in optical density during exposure is related to dose rate, as was seen for MD-55 film.[7] It is likely that the ΔOD increase between intra- and interexposure are indistinguishable at a low dose rate and therefore a separate radiation detector would be required to signal the end of exposure. This limiting dose rate may need to be established if the radiation sensitive medium of EBT film is to be used as a real-time *in vivo* dosimeter. The inset in Figure 4 illustrates ΔOD versus time for five pieces of film, each exposed to 9.52 Gy at 2.86 Gy/min. The measurements obtained for the same average dose rate is reproducible, yielding a 1.0% standard deviation in ΔOD at the end of exposure. This 1.0% deviation includes errors due to possible spatial variation in film response, which may be similar to that observed in MD-55,[12] measurement errors in the spectrophotometer results, light source intensity fluctuations, and deviations introduced due to variations in assembly of the film holder and phantom when replacing the film.[7]

It is interesting that ΔOD is slightly nonlinear with time (and hence with dose). This nonlinearity with dose is not unique to the real-time measurements reported here and can also be seen in the data provided by ISP, which were measured 1 h after exposure. It is not due to a shift in wavelength of maximum absorbance (λ_{max}), which was found to be quite stable over the entire 200 s exposure to 9.52 Gy (Fig. 5). The average λ_{max} for all exposures was calculated to be 635.6±0.7 nm (where the reported uncertainty is two times the standard deviation). The effect of changing the wavelength range of the spectral averaging window on ΔOD is shown in Fig. 6, illustrating that increasing the spectral range from 10 to 70 nm still showed a similar nonlinear effect, albeit less pronounced. One possibility is that the nonlinearity

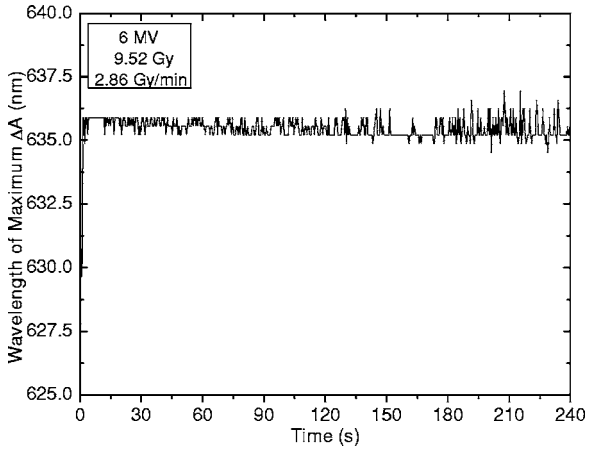

FIG. 5. Wavelength of maximum absorbance for EBT film vs. time during and after exposure to 9.52 Gy at 2.86 Gy/min with 6 MV x rays (beam on 0–200 s).

is either due to decrease in polymerization rate with dose, or due to saturation of carbon–carbon double bonds in the polymer. (The sensitive medium of GafChromic® EBT film is a modified version of that used in GafChromic® MD-55 and HS films.[9] The optical density of these films is based on the increase in number of chains with conjugated double and triple bonds that form during radiation induced polymerization.[7]) The ΔOD versus dose nonlinearity is not an issue for regular dosimetric use of this film, as a simple calibration plot is all that is required. The sensitive medium of this film may also be used for real-time *in vivo* dosimetry, provided that the total delivered dose is kept track of and the curves are insensitive to fluctuations in dose-rate and temperature that are typical of *in vivo* conditions. If so, a correction function would be needed to obtain a one-to-one correlation between ΔOD and dose. Otherwise, an appropriate model describing the polymerization kinetics, that can account and correct for all such variations, is desirable. A full understanding of chemistry and kinetics would require information on structure and packing of monomer units within the sensitive medium.

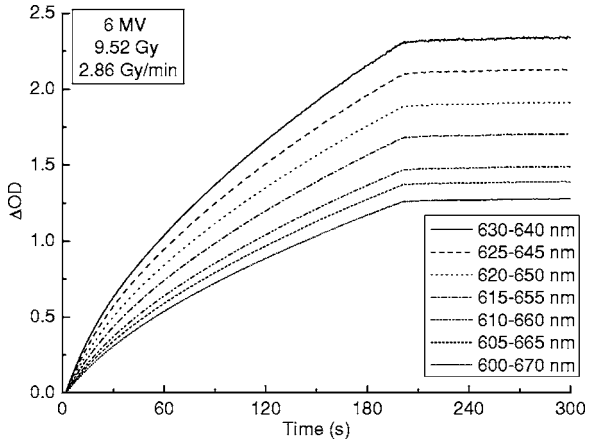

FIG. 6. Change in OD of EBT film vs. time for various spectral averaging windows.

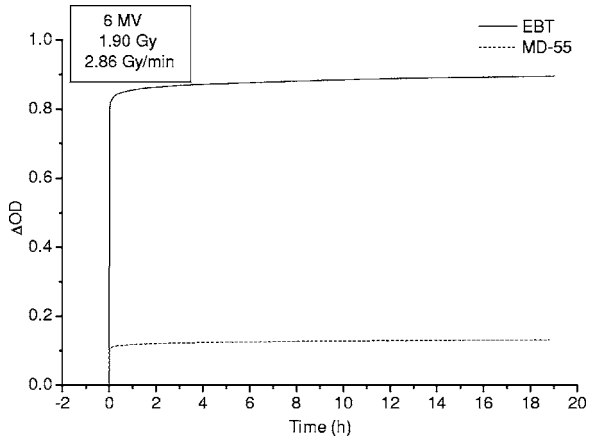

FIG. 7. Change in OD for EBT and MD-55 films during and after exposure.

The other possibility for the nonlinearity of ΔOD with dose is a change in the optical properties of the sensitive medium used in EBT film with dose. No information regarding these optical properties currently exists, and little can be deduced from a recent study of MD-55 film by Fusi *et al.*,[13] since the sensitive media in MD-55 and EBT films are different (see Sec. III D).

B. Sensitivity and stability comparison between EBT and MD-55 films

The ΔOD versus time for the two types of films due to a 1.9 Gy exposure with 6 MV x rays is shown in Fig. 7. The measurements were not normalized by thickness of radiation sensitive layer (each of EBT's radiation sensitive layers is 1 μm thicker than that of MD-55). Comparing or correcting for the thickness difference between MD-55 and EBT films is not particularly relevant; the two films have different radiation sensitive media, and their suspensions (both concentration and distribution) within the active layers are likely not the same. However, if a dosimeter were to be designed for a specific thickness, it would be useful in predicting the performance that could be achieved with each of these media.

Results show the EBT film to be 7.6±0.2 times (as measured immediately at the end of exposure) more sensitive than MD-55 film when exposed to approximately 2 Gy with 6 MV x rays. The sensitivity increase is defined here as the ratio of the optical densities for a given dose, although other definitions (such as the ratio of doses required to achieve a certain optical density) can be used and may give slightly different sensitivity value. Any discussion of sensitivity between MD-55 and EBT film depends on the energy at which the exposures to radiation are performed, since EBT is supposed to be more sensitive to low kilovolt energy photons than MD-55 (ISP product information). This makes EBT's response to ionizing radiation closer to that of water, potentially broadening the energy range over which it can be used as an *in vivo* dosimeter. Further, since optical density of EBT (measured near the absorbance peak) is not linear with dose, increase in sensitivity between EBT and MD-55 is also dose dependent for the definition used in this report.

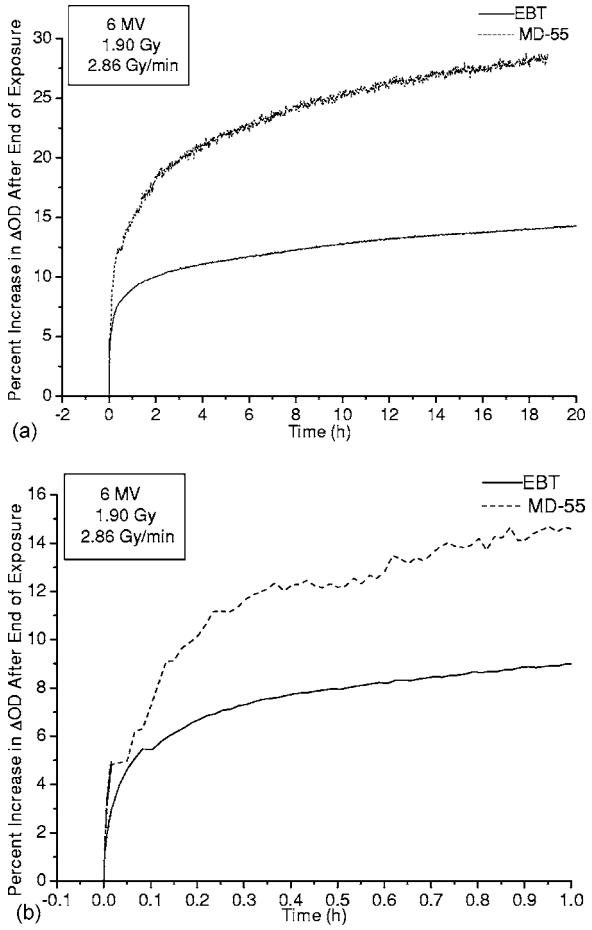

FIG. 8. (a) Percent increase in ΔOD for EBT and MD-55 films after exposure, calculated with respect to ΔOD at the end of exposure. (b) Percent increase in ΔOD for EBT and MD-55 films within 1 h after exposure.

The percent increase in ΔOD following exposure, calculated with respect to the value measured immediately at the end of exposure from the data of Fig. 7, is shown in Fig. 8(a). The percent increase for EBT film is nearly two times less than that of MD-55, and the signal begins to plateau earlier. At 18 h after exposure, the ΔOD of EBT film has increased by roughly 12.5%, whereas the increase for MD-55 film is more than 25%. The decreased postexposure darkening of EBT film, compared to that of MD-55 film, suggests that the polymerization kinetics occurring within the radiation sensitive medium are quicker, likely due to a different structure and packing of the sensitive material within the radiation sensitive layer (see Sec. III D). This is important for both conventional uses of these films and for the real-time dosimetry objectives in this work. Faster polymerization kinetics will allow accurate dose measurements soon after exposure without need for consideration of potential errors introduced by postexposure darkening. Similarly, accurate real-time dose measurements during the latter part of exposure will be possible without extraneous errors due to "postexposure" darkening of the dose delivered at the onset of exposure.

The percent increase in ΔOD reported here for MD-55 film are higher than was previously reported,[14,15] and will likely be higher than will be reported for EBT film. The reason is that other measurements compare the increase in optical density to the value measured soon after exposure, but not immediately after exposure as they often use a regular non-real-time densitometer. This data shows that a large portion of growth in ΔOD occurs within the first few minutes [Fig. 8(b)], and is thus not assessed in more conventional methods of measuring optical density for these films. It should be noted that the ΔOD drift values reported here are valid for the dose rate of 2.86 Gy/min, and may differ if another dose or dose rate is used.[3]

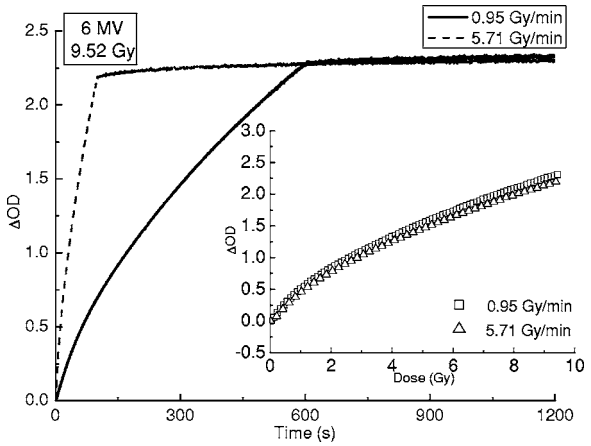

FIG. 9. Change in OD for EBT film exposed to 9.52 Gy, delivered with 6 MV at 0.95 and 5.71 Gy/min; change in OD normalized to dose during same exposure to 9.52 Gy (inset).

C. Dependence of real-time ΔOD measurements on dose rate for the EBT film

The change in OD versus time for a 9.52 Gy exposure delivered at two different dose rates (0.95 and 5.71 Gy/min) is shown in Fig. 9. If the film is used conventionally by measuring optical density some time after the end of exposure, the sixfold dose rate variation does not appear to introduce error, although further investigations to confirm this should be performed. With time axis converted to dose, the data obtained during exposure superimpose well (inset). However, analysis of variance of the ΔOD values as measured at the end of exposure revealed a statistical difference between the two groups, and hence a dose rate dependence for real-time measurements. The percent standard deviation (with respect to the mean) increased from 0.9% to 1.8% when both sets of data were included. Depending on the desired accuracy of dose estimate, this small dose rate dependence of real-time measurements may be tolerable. However, for the purposes of real-time *in vivo* dosimetry, dose and dose rate ranges over which dose rate variations do not introduce extraneous errors will need to be established. The ΔOD versus dose curve can be then used up to the established dose without correcting for dose rate dependent fluctuations. The dose rates used in this investigation (0.95 and 5.71 Gy/min) are applicable to external beam (linear accelerator output range of 100–600 MU/min, with treatments

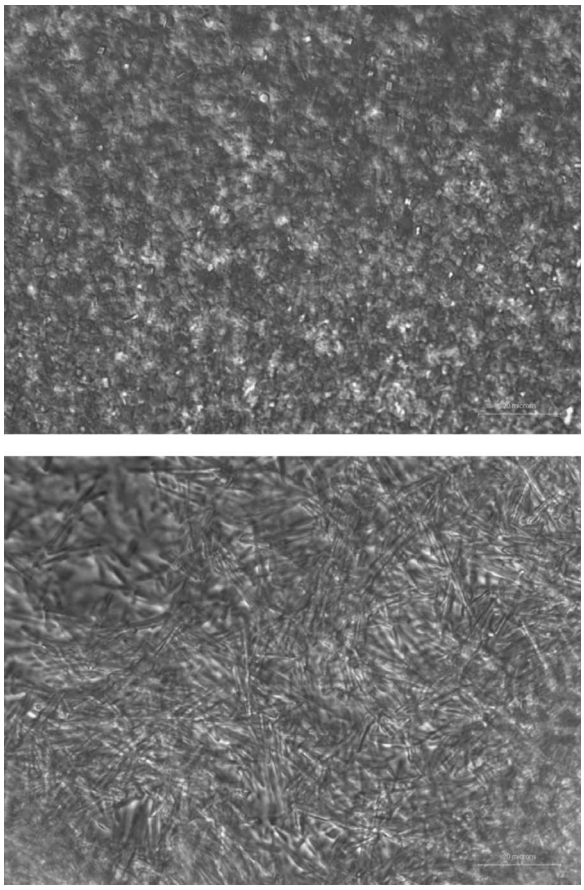

FIG. 10. Microscope images of monomer crystals within the sensitive media of MD-55 (top) and EBT (bottom) films (the blurring on the edges of the image is typical for an uneven thickness of sample).

typically delivered at 300 MU/min), and high dose rate brachytherapy.[16] To investigate whether this radiochromic medium is appropriate for low dose rate brachytherapy, dose rates down to a few cGy per hour will need to be used.[17]

D. Structure of active crystals in MD-55 and EBT films

Information about structure of a chemical compound can yield insight into its function. The microscope images of the sensitive media used in MD-55 and EBT films are shown in Fig. 10 (a 20 μm bar in bottom right corner indicates the scale). The monomer crystals used in MD-55 are sand-like (top image), and look similar as previously shown using scanning electron microscopy.[2] The monomer crystals within EBT, on the other hand, are elongated, stick-like structures (bottom image).

The active component used in EBT film is a modified version of that used in MD-55. Since three-dimensional structure and packing of diacetylene monomer crystals depends on the type and size of side groups,[18,19] it is reasonable to believe that the observed difference in the crystal structure is due to this type of modification. For MD-55 film, the polymerization reaction proceeds in only one direction with respect to the crystal axes,[20] and the absorbance is due to the conjugated double and triple bonds within the polymer chain.[21] The change in crystal structure with increased dose, evident through a shift in absorbance peak,[20,22]increases the separation between the last polymer unit and the next available monomer, decreasing the rate of polymerization.[9,7] Although a shift in λ_{max} is seen for MD-55,[1,5,14] no such shift in λ_{max} is seen for EBT film (Fig. 5). This suggests that the internal packing of the monomers within the active crystals of EBT is more stable, and that little rearrangement or separation occurs between the monomer and polymer structures. Thus no significant decrease in polymerization rate is expected to be seen. The decreased postexposure development observed for EBT film suggests that most of the polymerization occurs during exposure, and hence the polymerization rate does not in fact decrease with dose as much as for MD-55 film.

IV. CONCLUSION

The new GafChromic® EBT film is more sensitive to ionizing radiation dose and exhibits less postexposure darkening than GafChromic® MD-55 film. It shows a distinction between inter- and intraexposure rates of ΔOD increase, which allows easy identification of the end of exposure, but ΔOD during exposure is not linear with dose. Since λ_{max} is stable over the entire exposure to 9.52 Gy, increasing the spectral window of interest does not correct nonlinearity. This nonlinearity is thus likely due to the actual polymer chemistry that occurs in the sensitive medium of the film (such as depletion of monomers available for polymerization). It is not necessarily a deterrent from real-time dosimetry, as long as the correction function is stable against dose rate and temperature fluctuations. Under these conditions, a correction for this can be made using an appropriate model that can sufficiently describe the chemistry and kinetics during exposure. Developing such a model requires knowledge of monomer structure and packing of monomers within the sensitive medium. A small dose rate dependence in real-time measurements, yielding an increase in standard deviation from 0.9% to 1.8%, is seen for a dose of 9.52 Gy and a dose rate range between 0.95 and 5.71 Gy/min. For EBT film to be useful in real-time *in vivo* dosimetry, a dose and dose rate ranges, at which dose rate dependent errors are not seen, need to be established. Dependence of ΔOD measurements and λ_{max} on temperature will also need to be investigated. Although further investigations are required, GafChromic® EBT's increase in sensitivity and rates of polymerization make it a promising medium for real-time *in vivo* dosimetry.

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