

UNLAMINATED GAFCHROMIC EBT3 FILM FOR ULTRAVIOLET RADIATION MONITORING

David Welch^{1,*}, Gerhard Randers-Pehrson¹, Henry M. Spotnitz² and David J. Brenner¹
¹Center for Radiological Research, Columbia University Medical Center, New York, NY, USA
²Department of Surgery, Columbia University Medical Center, New York, NY, USA

*Corresponding author: dw2600@columbia.edu

Received 29 December 2016; revised 25 January 2017; editorial decision 27 January 2017;
accepted 29 January 2017

Measurement of ultraviolet (UV) radiation is important for human health, especially with the expanded usage of short wavelength UV for sterilization purposes. This work examines un laminated Gafchromic EBT3 film for UV radiation monitoring. The authors exposed the film to select wavelengths in the UV spectrum, ranging from 207 to 328 nm, and measured the change in optical density. The response of the film is wavelength dependent, and of the wavelengths tested, the film was most sensitive to 254 nm light, with measurable values as low as 10 $\mu\text{J}/\text{cm}^2$. The film shows a dose-dependent response that extends over more than four orders of magnitude. The response of the film to short wavelength UV is comparable to the daily safe exposure limits for humans, thus making it valuable as a tool for passive UV radiation monitoring.

INTRODUCTION

Radiation from the ultraviolet (UV) range of the electromagnetic spectrum is involved in many aspects of human health with both beneficial and harmful effects^(1, 2). The UV wavelengths are commonly divided into four regions: vacuum UV (10–200 nm), short-wave UV (UVC, 200–280 nm), medium-wave UV (UVB, 280–315 nm) and long-wave UV (UVA, 315–400 nm). Vacuum UV can generally be ignored with regard to human health because it cannot propagate through air since it is absorbed by molecular oxygen. The distinctions within the remainder of the UV range (UVC, UVB and UVA) are dependent on their phenomenological effects. UVC, which is not present in solar radiation because it is unable to penetrate the stratosphere of the earth, can cause photokeratitis⁽³⁾, cataracts^(4, 5) and skin cancer⁽⁶⁾, but it is also effective at killing single-celled organisms. The UVB range is instrumental to the synthesis of Vitamin D in human skin while concurrently being the cause of sunburns and skin cancer. UVA can cause tanning and redness similar to UVB but requires much larger amounts of energy to produce an effect. Accurate dosimetry of solar UV radiation in the UVA and UVB ranges has been an area of research for many years⁽⁷⁾. As artificially generated UVC has gained more prominence, particularly due to germicidal sterilization at 254 nm of both airborne and waterborne pathogens^(8–11), dosimetry in this region has also become increasingly important. Furthermore, the expanded use of other wavelengths of UVC, such as our recent work with 207 nm light that has shown promise as a new means of sterilization safe from negative biological

effects^(12, 13), calls for continued improvement of dosimetry across the spectrum.

Measurement of UV radiation can be performed with physical, biological or chemical means⁽⁷⁾. Physical measurements quantify either thermal power, with devices such as thermopiles or pyroelectric radiometers, or photon detection, which is used in photomultiplier tubes and photodiodes. Biological dosimetry can indicate UV radiation through the inactivation of cells or viruses or even simply through the degree of erythema on human skin. Chemical measurements can take place in solids, liquids or gases and measure through integration of radiant power received over an exposure period. The most common chemical measurement devices take the form of film. Specialized films exposed to radiation undergo chemical reactions which, either directly or with subsequent processing, lead to an absorption change in the UV or visible range. Diazo films are primarily sensitive to UVA and necessitate development with ammonia vapor⁽¹⁴⁾. Polysulphone films show a relative sensitivity close to human skin but require measuring absorbance changes in the UV region to quantify results^(15, 16). Other work has been performed which showed various UV sensitive drugs or dyes could be incorporated into polyvinyl chloride film^(17–19) or polyvinyl butyral^(20, 21).

Recently, the primary film studied for use in UV dosimetry has been commercially available radiochromic film^(22–25). Most of these works have focused on solar radiation monitoring, i.e. UVA and UVB only. The exception is the work by Aydarous *et al.* which, in addition to solar radiation monitoring,

claimed to measure into the UVC range with Gafchromic EBT3⁽²²⁾. However, two factors call into question the validity of the conclusions Aydarous *et al.* put forward, specifically regarding UVC dosimetry. The first factor is that the light source used (UVLMS-38, UVP LLC, Upland CA) is not well filtered at the 254 nm illumination setting. Obtaining the output spectra through communication with the vendor, UVP LLC, revealed that while the 254 nm peak is dominant there are also significant power contributions from peaks at 313 and 365 nm. The second factor is that polyester, which sandwiches the UV sensitive active region of the film, almost completely blocks transmission of wavelengths shorter than ~320 nm^(26, 27). These two factors together suggest that Aydarous *et al.* were actually continuing to measure UVA and UVB exposure when they claimed to measure UVC.

In this work, a special variation of Gafchromic EBT3 film for UV radiation measurement was examined. This new film, referred to as unlaminated Gafchromic EBT3, lacks the polyester laminate on one side thus exposing the active region. With the active region exposed, the film is much more sensitive to UVC and thus can serve as a radiation measurement device.

MATERIALS AND METHODS

Light characterization

The spectrum of each UV light source was measured with a spectrometer (SPM-002-BT, Photon Control,

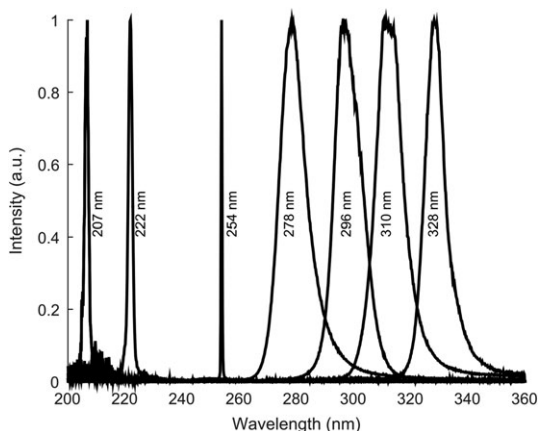


Figure 1. Radiometric spectra for each of the light sources used in this study are shown normalized to their peak intensity (expressed in arbitrary units). The 207, 222, 254 and 296 nm sources utilized bandpass filters centered at the respective wavelengths. The three solid-state UV emitters (278, 310 and 328 nm) primarily emit at a single wavelength thus no extra filtering was used.

Inc., Burnaby, BC), which was radiometrically calibrated with a deuterium lamp (63 945, Newport, Irvine, CA). Optical power measurements were performed using an 818-UV/DB low-power UV enhanced silicon photodetector with an 843-R optical power meter (Newport, Irvine, CA).

Light sources

Various sources and filters to generate and isolate specific wavelengths were used in this study. An excimer lamp (High Current Electronics Institute, Tomsk, Russia) with a krypton–bromine (Kr–Br) gas mixture to principally emit at 207 nm was used. A custom bandpass filter (208NB6, Omega Optical, Brattleboro, VT), with center wavelength (CWL) of 208^{+2}_{-1} nm and full width at half maximum (FWHM) of 6^{+2}_{-1} nm, was used to essentially remove all but the dominant 207 nm emission. Similarly, a separate excimer lamp (High Current Electronics Institute, Tomsk, Russia) with a krypton–chlorine (Kr–Cl) gas mixture was used to emit principally at 222 nm. A custom bandpass filter (224NB7, Omega Optical, Brattleboro, VT), CWL of 224^{+2}_{-1} nm and FWHM of 7^{+2}_{-1} nm, was used with the Kr–Cl lamp. For 254 nm, a Mineralight XX-15S UV Bench Lamp (UVP, Upland, CA) with a mercury line filter (10MLF10-254, Newport, Irvine, CA) with a CWL of 253.7 ± 3 nm and FWHM of 11 ± 3 nm was used. For 296 nm, a deuterium lamp (63 945, Newport, Irvine, CA) with a bandpass filter (296BP8, Omega Optical, Brattleboro, VT) with CWL 296 ± 1.5 nm and FWHM of 8 ± 2 nm was used.

The remaining wavelengths included in this investigation were produced with solid-state UV emitters. The authors found that the emitter designed for a peak output at 280 nm (MTE280F13-UV, Marktech Optoelectronics, Latham, NY) had a CWL of 278 nm with a FWHM of 10 nm. The emitter designed for 310 nm (MTE310F13-UV, Marktech Optoelectronics) had a CWL of 310 nm with a FWHM of 10 nm. The emitter designed for 325 nm (MTE325F13-UV, Marktech Optoelectronics) had a CWL of 328 nm with a FWHM of 9 nm. The variations in CWL for the nominally 280 and 325 nm emitters were within the tolerance stated by the manufacturer.

Figure 1 shows the radiometric spectra for all of the sources used in this study. Each spectrum was normalized to the intensity value at the CWL.

Film structure

The film used in this study was a specialty order from Ashland Specialty Ingredients (Bridgewater, NJ) referred to as unlaminated Gafchromic EBT3 (Product Code 849 952). This specialty film is essentially one half of the regular Gafchromic EBT3 film.

Regular EBT3 film comprises an active layer, nominally 28 μm thick, sandwiched between two 125 μm matte-polyester substrates. The active layer contains a proprietary mixture of active component, marker dye, stabilizers and other components. The unlaminate EBT3 film used in this study is simply a 14 μm thick active layer on top of a single 125 μm polyester substrate.

Film analysis

Films were scanned as 48 bit RGB TIFF images using an Epson Perfection V700 Photo flatbed scanner (Epson, Suwa, NGN, Japan). The optical density values for each image file were extracted using custom software developed by Alves *et al.*⁽²⁸⁾. The authors chose to evaluate the optical density (OD) solely in the red channel because it has been shown to have the highest dose-dependent response^(29, 30). The background signal from an unexposed piece of film was subtracted to give the Net OD using the following equation:

$$\text{Net OD} = \text{OD} - \text{OD}_{\text{blank}} = d_x(D), \quad (1)$$

which is also the dose response, d_x , for a given dose (D). Data for each wavelength were matched to a fitting function with the form

$$d_x(D) = \frac{a + bD}{D + c}, \quad (2)$$

where a , b and c are constants. The fitting function was optimized in MATLAB (MathWorks, Natick, MA) using the curve fitting tool to minimize the squared difference between the experimental data and the fit equation.

Test of regular EBT3 response to 254 nm

Because the 254 nm UV source Aydarous *et al.* used was not well filtered, the exposure testing of standard EBT3 film was replicated. The authors exposed a piece of Gafchromic EBT3 film (Product Code 828 204, Ashland Specialty Ingredients) to the 254 nm filtered source for a total exposure of 5 J/cm². The film was analyzed for Net OD change against a piece of unexposed EBT3 film.

RESULTS

Film response

The relationship between exposure and Net OD for each exposure wavelength band is shown in Figure 2. Each wavelength band evaluated shows the same general response curve shape but the response shows a wavelength-dependent shift.

Fitting function

A fitting function, matching the form of Equation (2), was determined for each of the wavelength response curves in Figure 2. Table 1 gives the constant values for each fit equation. The coefficient of determination value, denoted as R^2 , is also given to indicate how well the model replicates the observed outcomes.

Relative spectral effectiveness

The American Conference of Governmental Industrial Hygienists (ACGIH) has established public health standards for incoherent UV radiation⁽³¹⁾. Threshold limit values (TLVs) represent the conditions under which it is believed that nearly all healthy workers may be exposed without adverse health effects such as erythema and photokeratitis. The ACGIH recommends that the total daily exposure to UV is dose limited to 3 mJ/cm². The TLVs for UV radiation are wavelength dependent with the peak spectral effectiveness at 270 nm. Daily exposure limits are therefore best expressed as relative spectral effectiveness values ($S(\lambda)$) referred to as the hazard function, where $S(\lambda)$ is normalized to a value of 1 at 270 nm. A plot of the ACGIH UV radiation hazard function is shown in Figure 3.

Radiation monitors in the UV range are often compared against the hazard function to assess their fitness for human hazard monitoring⁽⁷⁾. The authors have plotted the relative sensitivity of the film assessed in this work by using the fit equations to determine the exposure necessary to achieve a Net OD response of 0.4, taking the inverse of the exposure value, and then normalizing the values so that the response of this film at 254 nm aligns with the hazard function value. These data points are also plotted in Figure 3.

Test of EBT3 response to 254 nm

A Net OD change of 0.010 when exposing regular EBT3 to 5 J/cm² at 254 nm was observed. This is in contrast to the Net OD of 0.125 Aydarous *et al.* measured for their exposure.

DISCUSSION

The characterization of unlaminate EBT3 film demonstrates its utility as a UV radiation measurement device. The film is sensitive to a wide range of UV wavelengths and the response for a given exposure is strongly wavelength dependent. A peak sensitivity using a 254 nm light source was observed. Each wavelength tested in this study showed a similar shape in its response curve, plotted in Figure 2. This similarity suggests that the film could be used for measurement of an exposure at any wavelength throughout UVB and UVC given a calibration curve

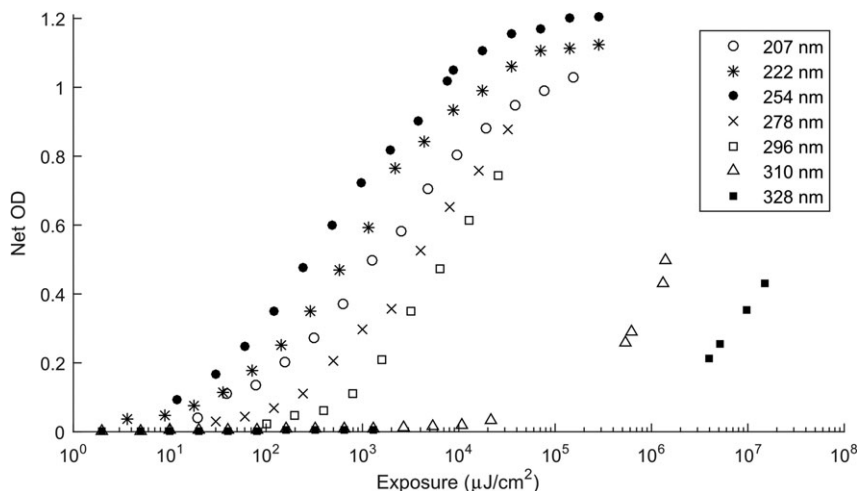


Figure 2. The film response (Net OD) for increasing exposures is plotted for each of the wavelengths tested. The film shows a similar response profile shape, which shifts dependent on the wavelength.

Table 1. Values of constants used in Equation (2) to define the fitting function for each wavelength.

Wavelength (nm)	a	b	c	R^2
207	180	0.986	1790	0.988
222	76.1	1.08	997	0.992
254	113	1.15	723	0.983
278	146	0.925	3220	0.992
296	39.3	0.891	5480	0.999
310	1.34×10^4	0.996	1.58×10^6	0.996
328	1.64×10^4	0.629	7.45×10^6	0.999

The rightmost column gives the coefficient of determination for each fit equation.

is performed. The fitting functions calculated in this work show very good agreement to the experimental results, with R^2 values close to 1.0, so they can be applied to interpolate any exposure given the wavelength and a Net OD within the calibration range.

This film has many of the advantages of the regular version of EBT3: it is flexible, develops in real time without post-exposure treatment, has a high spatial resolution and experiences a color change in the visible range so it is easily analyzed with a commercial optical scanner. Unlaminated EBT3 also has a wide dynamic range. The authors demonstrate measuring exposures at 254 nm as low as $10 \mu\text{J}/\text{cm}^2$ and as high $100 \text{ mJ}/\text{cm}^2$. While the exact bounds of the sensitivity range are wavelength dependent, the width of the range appears to remain constant for different wavelengths.

Unlike regular EBT3, which has a symmetrical structure and can be radiated from any directions using energies from keV to MeV, the unlaminated film does have a directional dependency because the polyester backing blocks much of the UVB and UVC light from reaching the active layer. When the film is radiated on the side with the active region, it exhibits a nearly ideal cosine response equivalent to other radiochromic films. The absence of the polyester laminate over the active region does leave the active region susceptible to removal since it is water soluble, but this can be avoided with minimal handling precautions. Otherwise, the same care and handling afforded to regular EBT3 films should be applied to this unlaminated film variation.

Some of the sources used in this study, in particular the 296 nm filtered source and the solid-state emitters, have FWHM bandwidths of up to 10 nm. As pointed out in the work by Chaney and Sliney, these large bandwidths can introduce errors in the evaluation of spectral effectiveness, particularly in the regions where the slope is very high⁽³²⁾. Uncertainties in the spectral effectiveness in UVB can arise because a large slope gives disproportionately higher weight to the lower wavelengths within each bandwidth. A more thorough assessment of response in the UVB region, where a large drop in the spectral effectiveness occurs, could be performed with the use of a monochromator with a narrow bandwidth. unlaminated EBT3 shows a deviation from the hazard function at 280 nm, which is before the region of the hazard function with the highest slope, thus this source of

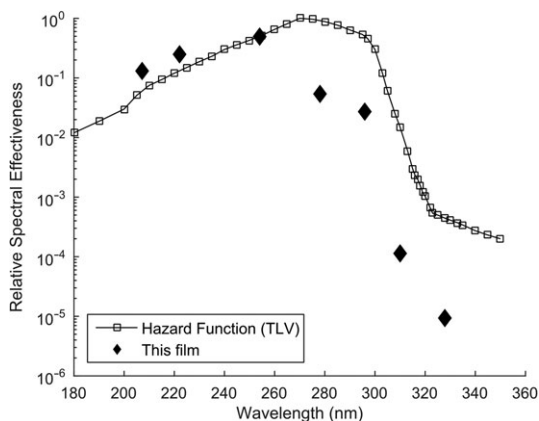


Figure 3. The relative spectral effectiveness ($S(\lambda)$) of the TLVs published by the ACGIH, known as the hazard function, is plotted as a function of wavelength in the UV range⁽³¹⁾. The hazard function is simply TLVs normalized to their maximum value at 270 nm. The relative spectral effectiveness values of the film tested in this study have been normalized to match the effectiveness of the hazard function at 254 nm, which is 0.5.

uncertainty will likely have no influence on the assessment of this film for overall radiation monitoring.

While Aydarous *et al.* alleged to measure UVC light with regular EBT3 film, the authors believe errors in their work invalidate their results. The test of regular EBT3 with a properly filtered 254 nm source showed a Net OD change of only ~8% of their measured value. The disparity between the results is likely attributed to proper experimental irradiance procedures. Regular EBT3 does experience a color change, demonstrated here with an exposure of 5 J/cm² of 254 nm UVC, but the sensitivity range is vastly different than un laminated EBT3 film; this discrepancy can be attributed to the attenuation through the polyester.

The authors have demonstrated that the un laminated EBT3 film presented here accurately measures throughout the UV spectrum. Furthermore, the sensitivity is such that it aligns well with the TLVs in the UVC range. For example, the TLV at 254 nm is 6.0 mJ/cm² and thus centered within the sensitivity range of this film. This alignment means that the film could be employed as a safety monitor given the spectrum of UV exposure was known so it could be correctly calibrated. This film would not be adequate for overall UV hazard assessment because its spectral response does not align well with the hazard curve, as shown in Figure 3. However, if exposure conditions are well characterized, such as situations using filtered sources, this film would be an excellent choice as a passive UV radiation monitor.

CONCLUSION

The un laminated EBT3 film examined in this work has a number of key attributes that make it useful for UV radiation measurement. The film has a high sensitivity, with exposures in the $\mu\text{J}/\text{cm}^2$ range detectable for much of the of the UVC range. The film also has a large dynamic range, with exposure measurements across more than four orders of magnitude possible. Because this film responds by the same means as regular EBT3 film, the processing and subsequent dose measurement are straightforward. The response can be fit to an easily invertible function and thus precise exposure levels are simple to obtain from only a few calibration points. The film shows a response in the UVC range that, although not exactly aligned with the hazard function, is within the same exposure range important for human health studies. Overall, the authors have shown that this commercially available film variant can be a valuable tool for quick, reliable and accurate assessment of UV radiation, especially in the UVC range.

FUNDING

This work was supported by Columbia-Coulter Translational Research Partnership 9/1/15–8/31/16 ‘Ultraviolet Laser Sleeve’ grant to Drs Spotnitz and Andreas Hielscher.

REFERENCES

1. Parrish, J. A., Anderson, R. R., Urbach, F. and Pitts, D. UV-A Biological Effects of Ultraviolet Radiation with Emphasis on Human Responses to Longwave Ultraviolet (Springer US) (1978) ISBN 978-1-4684-2477-5.
2. Sliney, D. H. *Ultraviolet radiation exposure criteria*. Radiat. Prot. Dosim. **91**(1-3), 213–222 (2000).
3. Sliney, D. H. *Ultraviolet radiation effects upon the eye: problems of dosimetry*. Radiat. Prot. Dosim. **72**(3-4), 197–206 (1997).
4. Jose, J. G. and Pitts, D. G. *Wavelength dependency of cataracts in albino mice following chronic exposure*. Exp. Eye Res. **41**(4), 545–563 (1985).
5. Söderberg, P. G. *Acute cataract in the rat after exposure to radiation in the 300 nm wavelength region A study of the macro-, micro- and ultrastructure*. Acta Ophthalmol. (Copenh). **66**(2), 141–152 (1988).
6. Pfeifer, G. P. and Besaratinia, A. *UV wavelength-dependent DNA damage and human non-melanoma and melanoma skin cancer*. Photochem. Photobiol. Sci. **11**(1), 90–97 (2012).
7. Diffey, B. *Ultraviolet Radiation Dosimetry and Measurement* (Boston, MA: Springer US) (1986) ISBN 978-1-4899-0571-0.
8. Rahn, R. O. *Dosimetry of room-air germicidal (254 nm) radiation using spherical actinometry*. Photochem. Photobiol. **70**(3), 314–318 (1999).

9. Glaze, W. H., Kang, J.-W. and Chapin, D. H. *The chemistry of water treatment processes involving ozone, hydrogen peroxide and ultraviolet radiation*. *Ozone Sci. Eng.* **9**(4), 335–352 (1987).
10. Yin, R., Dai, T., Avci, P., Jorge, A. E. S., de Melo, W. C. M. A., Vecchio, D., Huang, Y.-Y., Gupta, A. and Hamblin, M. R. *Light based anti-infectives: ultraviolet C irradiation, photodynamic therapy, blue light, and beyond*. *Curr. Opin. Pharmacol.* **13**(5), 731–762 (2013).
11. McDevitt, J. J., Rudnick, S. N. and Radonovich, L. J. *Aerosol susceptibility of influenza virus to UV-C light*. *Appl. Environ. Microbiol.* **78**(6), 1666–1669 (2012).
12. Buonanno, M., Randers-Pehrson, G., Bigelow, A. W., Trivedi, S., Lowy, F. D., Spotnitz, H. M., Hammer, S. M. and Brenner, D. J. *207-nm UV light – a promising tool for safe low-cost reduction of surgical site infections. I. In vitro studies*. *PLoS One* **8**(10), e76968 (2013).
13. Buonanno, M., Stanislauskas, M., Ponnaiya, B., Bigelow, A. W., Randers-Pehrson, G., Xu, Y., Shuryak, I., Smilenov, L., Owens, D. M. and Brenner, D. J. *207-nm UV light—a promising tool for safe low-cost reduction of surgical site infections. II: In-vivo safety studies*. *PLoS One* **11**(6), e0138418 (2016).
14. Ali, J. B. and Jacobson, R. E. *The use of diazo film as a film badge dosimeter*. *J. Photograph. Sci.* **28**(4), 172–176 (1980).
15. Davis, A., Deane, G. H. W. and Diffey, B. L. *Possible dosimeter for ultraviolet radiation*. *Nature* **261**(5556), 169–170 (1976).
16. Davis, A., Diffey, B. L. and Tate, T. K. *A personal dosimeter for biologically effective solar UV-B radiation*. *Photochem. Photobiol.* **34**(2), 283–286 (1981).
17. Diffey, B. L. and Davis, A. *A new dosimeter for the measurement of natural ultraviolet radiation in the study of photodermatoses and drug photosensitivity*. *Phys. Med. Biol.* **23**(2), 318 (1978).
18. Abdel-Rehim, F., Ebrahim, S. and Abdel-Fattah, A. A. *The suitability of a dyed plastic film for biologically effective UVB radiation measurement*. *J. Photochem. Photobiol. A Chem.* **73**(3), 247–251 (1993).
19. Ebraheem, S., Abdel-Fattah, A. A., Said, F. I. and Ali, Z. I. *Polymer-based triphenyl tetrazolium chloride films for ultraviolet radiation monitoring*. *Radiat. Phys. Chem.* **57**(2), 195–202 (2000).
20. Abdel Rehim, F., Basfar, A. A. and Abdel-Fattah, A. *The use of Riso B3 film gamma dosimeter for monitoring ultraviolet radiation*. *J. Photochem. Photobiol. A Chem.* **101**(1), 63–67 (1996).
21. Abdel-Fattah, A. A., El-Kelany, M., Abdel-Rehim, F. and El Miligy, A. A. *UV-sensitive indicators based on bromophenol blue and chloral hydrate dyed poly(vinyl butyral)*. *J. Photochem. Photobiol. A Chem.* **110**(3), 291–297 (1997).
22. Aydarous, A., Al-Omary, E. A. and El Ghazaly, M. *Characterization of Gafchromic EBT3 films for ultraviolet radiation dosimetry*. *Radiat. Effect. Defect. Solids* **169**(3), 249–255 (2014).
23. Chun, S. L. and Yu, P. K. N. *Note: calibration of EBT3 radiochromic film for measuring solar ultraviolet radiation*. *Rev. Sci. Instrum.* **85**(10), 106103 (2014).
24. Butson, E. T., Cheung, T., Yu, P. K. N. and Butson, M. J. *Measuring solar UV radiation with EBT radiochromic film*. *Phys. Med. Biol.* **55**(20), N487 (2010).
25. Butson, M. J., Cheung, T., Yu, P. K. N., Abbati, D. and Greenoak, G. E. *Ultraviolet radiation dosimetry with radiochromic film*. *Phys. Med. Biol.* **45**(7), 1863 (2000).
26. Cybulski, W. J. and Peterjohn, W. T. *Effects of ambient UV-B radiation on the above-ground biomass of seven temperate-zone plant species*. *Plant Ecol.* **145**(1), 175–181 (1999).
27. Kieber, D. J., Miller, G. W., Neale, P. J. and Mopper, K. *Wavelength and temperature-dependent apparent quantum yields for photochemical formation of hydrogen peroxide in seawater*. *Environ. Sci. Process. Impacts* **16**(4), 777–791 (2014).
28. Alves, V. G. L., Cardoso, S. C. and da Silva, A. X. *Gafchromic EBT2 dosimetry via robust optimization*. *Comput. Phys. Commun.* **184**(7), 1708–1716 (2013).
29. Micke, A., Lewis, D. F. and Yu, X. *Multichannel film dosimetry with nonuniformity correction*. *Med. Phys.* **38**(5), 2523–2534 (2011).
30. Devic, S., Tomic, N., Soares, C. G. and Podgorsak, E. B. *Optimizing the dynamic range extension of a radiochromic film dosimetry system*. *Med. Phys.* **36**(2), 429–437 (2009).
31. *Threshold Limit Values and Biological Exposure Indices*. American Conference of Governmental Industrial Hygienists (2012) ISBN 978-1-607260-48-6.
32. Chaney, E. K. and Sliney, D. H. *Re-evaluation of the ultraviolet hazard action spectrum—the impact of spectral bandwidth*. *Health Phys.* **89**(4), 322–332 (2005).