Received:
12 January 2015

Revised:
3 August 2015

Accepted:
5 October 2015

doi: 10.1259/bjr.20150035

Cite this article as:

Mukherjee B, Gholami YH, Bhonsle U, Hentschel R, Khachan J. A unique alpha dosimetry technique using Gafchromic EBT3® film and feasibility study for an activity calibrator for alpha-emitting radiopharmaceuticals. *Br J Radiol* 2015; **88**: 20150035.

SHORT COMMUNICATION

A unique alpha dosimetry technique using Gafchromic EBT3® film and feasibility study for an activity calibrator for alpha-emitting radiopharmaceuticals

^{1,2}BHASKAR MUKHERJEE, Dr.rer.nat, ²YASER H GHOLAMI, MSc, ³UDAY BHONSLE, MSc, ¹REINHARD HENTSCHEL, Dr.Ing and ²JOSEPH KHACHAN, PhD

¹Detector and Sensor Group, West German Proton Therapy Centre Essen (WPE), Essen, Germany

²Department of Medical Physics, School of Physics (A28), The University of Sydney, Sydney, NSW, Australia

³Division of Physical and Chemical Sciences, International Atomic Energy Agency (IAEA), Vienna, Austria

Address correspondence to: Dr Bhaskar Mukherjee

E-mail: bhamukh@gmail.com

Objective: To develop an alpha dosimetry technique for activity calibration of alpha-emitting radiopharmaceuticals using the Gafchromic® EBT3 (Gaf-EBT3) radiochromic film (International Speciality product, Wayne, NJ).

Methods: The Gaf-EBT3 has a tissue equivalent radiosensitive layer (approximately 28 µm) sandwiched between two 100-µm thick polyester sheaths, thereby making it insensitive to alpha particles. We have split a Gaf-EBT3 sheet using a surgical scalpel to remove one of the polyester protective layers and covered the radiosensitive layer with thin Mylar® foil (Goodfellow Cambridge Limited, Huntingdon, UK) (2.5 µm). Small pieces of modified film were exposed at contact with a 560-Bq thin ²⁴¹Am source for 5, 10, 24 and 94 h. The optical density of the films was evaluated using an optical densitometer. The alpha energy spectra of the ²⁴¹Am source were recorded using a Si(Li) surface barrier detector.

Results: Time-integrated specific alpha surface activity (kBq cm⁻² h) was represented as a function of optical density.

Conclusion: By removing one of the 100 µm thick polyester protective layers, the authors have modified the Gaf-EBT3 film to a sensitive alpha dosimeter. The calibration function relevant to a ²⁴¹Am reference source was evaluated from the optical densities of the dosimeter foils. Furthermore, calibration functions for important alpha emitters such as ²²³Ra, ²²⁵Ac or ²¹⁰Pb were parameterized from the ²⁴¹Am reference data.

Advances in knowledge: The authors have developed and tested the principle of a clinical alpha dosimeter using Gaf-EBT3 radiochromic films originally developed for photon dosimetry. This novel, user-friendly technique could be implemented in quality assurance and calibration procedures of important alpha-emitting radiopharmaceuticals prior to their clinical applications.

INTRODUCTION

The novel developments of radionuclide production and advances in targeted internal therapy (TIT) have promoted a particular interest in alpha-particle emitters for the treatment of micrometastatic diseases.¹ The usual external beam radiation therapy is a mainstay of cancer treatment. However, a major drawback is the lack of specificity, which can result in normal tissue toxicity. TIT is an alternative approach, which can seek molecular targets within a tumour site and allows a sensitive identification between the normal and tumour cells. There are different alpha-emitting isotopes available for human clinical use such as ²¹¹At, ²¹²Pb, ²²⁵Ac, ²²³Ra and ¹⁴⁹Tb. These isotopes emit alpha particles within the energy range of 4–8 MeV.² Currently, ²²³Ra is the most promising radionuclide that can be used for treating bone metastases associated with prostate cancer.^{1,3} Large quantities of ²²³Ra with half-life

of 11.4 days can be acquired from uranium mill tailing. It could also be obtained as a generator based on the ²²⁷Ac parent with half-life of 21.8 years.⁴ Alpharadin (²²³Ra chloride) is an alpha-emitting radioisotope recently approved by the Food and Drug Administration to treat bone metastases associated with prostate cancer. ²²³Ra has a half-life of 11.4 days, and it basically emits four alpha particles per decay, which is 95% of the total energy emitted and about <2% of the energy originates from photons which could be used for qualitative imaging.^{3,5} There have been suggestions to customize these alpha-emitting radionuclides for treating different types of cancers by enclosing them in a nanoparticle cage and linking the antibodies on their surface.⁶

Evidently, suitable calibration and quality assurance (QA) procedures are prerequisite for clinical trials and routine

Figure 1. Schematic diagram showing the cross-section of the EBT3 radiochromic film widely used in photon dosimetry applications. The splitting of the sensitive dye layer from protective polyester sheath using a surgical scalpel is highlighted.

applications of the above radiopharmaceuticals. Most of the alpha radionuclides co-emit low-intensity characteristic γ -rays. Gamma spectrometers are used to assess those γ -rays for the purpose of calibration. However, modern high-resolution gamma spectrometers are bulky and expensive. This motivated the authors to develop a user-friendly calibration method based on alpha dosimetry utilizing widely used radiochromic films.

Radiochromic films belong to the category of chemical dose-meters. The principle is based on the phenomenon of permanent colouration of a thin radiosensitive layer (wrapped with transparent polyester foils) caused by ionizing radiations.⁷ Unlike photographic films, radiochromic films require no chemical processing and are now widely used in medical radiation dosimetry.⁸ Alpha particles possess very short range in materials and, hence, are completely stopped by the protective polyester sheaths before reaching the radiosensitive layer. This makes common radiochromic films insensitive to alpha particles.

The authors have overcome this drawback by splitting the radiochromic film, removing one of the 100 μm (thick) protective polyester layers and replacing it with 2.5- μm (thin) Mylar® foil (scientific name: polyethylene terephthalate; specification: ES301025; supplier: Goodfellow Cambridge Ltd,

Figure 2. Showing the normalized spectrum of the 560-Bq ^{241}Am alpha source recorded with a high-resolution Si(Li) surface barrier detector. The unattenuated spectrum and the spectrum attenuated using a 2.5 μm Mylar foils are shown by “filled circles” and “hollow circles”, respectively. One observes the main energy peak shifted from 5.48 to 5.21 MeV owing to energy loss in the Mylar foil. Consequently, the width (full width at half maximum) of main peak increased from 26 to 45 keV.

Figure 3. Photograph of the modified EBT3 film pieces irradiated with alpha particles from a 560-Bq ^{241}Am source for: (a) 5 h, (b) 10 h, (c) 24 h and (d) 94 h. The rectangular film sections are the unexposed samples cut out from the same lot used for alpha irradiation. These specimens were used as references (control) for optical density calculation.

Huntingdon, UK). This facilitates alpha particles to reach the sensitive layer without significant energy loss. Based on this principle, we have developed an alpha dosimeter suitable for versatile applications including calibration of alpha-emitting radiopharmaceuticals.

METHODS AND MATERIALS

Film sample preparation

The most recent brand of radiochromic film (Gafchromic® EBT3; International Speciality Products, Wayne, NJ) was used in this study. The EBT3 sheet (20.3×25.4 cm) is made of an approximately 28- μm tissue equivalent radiosensitive active dye layer sandwiched between two 100- μm polyester protective foils. Radiochromic films are principally sensitive to photons and are widely used by medical and radiological physics communities.^{7,8} The radiosensitive dye layer and protective foils were not firmly glued to each other. Therefore, one of the protective foils could be conveniently separated using a splitting blade (surgical scalpel) as depicted in Figure 1. This allows alpha particles to interact directly with the dye layer. Two square (5×5 cm) sections were cut out from a single EBT3 film sheet, with one protective layer removed. The dimensions of the sections were evaluated using a digital calliper (Oehmen Labortechnik, Cologne,

Figure 4. Schematic diagram of the modified Gafchromic® EBT3 film cross-section used in alpha depth-dose calculations. E_0 represents the unattenuated alpha energy from the source ($d = 0.7$ cm). E_1 and E_2 represent the energy of the alpha particles entering the dye layer ($T_D = 23$ μm) after passing through the Mylar foil ($T_M = 2.5$ μm) and exit energy, respectively. ΔE stands for the alpha energy loss in the radiosensitive dye layer.

Figure 5. Depicting the range of alpha particles as functions of energy (a) and subsequently alpha energy as functions range (b) in Mylar and EBT3 dye (tissue equivalent) layer. Data points were adopted from the literature¹¹ and fitted with polynomials (Table 1).

Germany) with an accuracy of ± 0.01 mm. The first section (area = 25 cm^2) was covered with $2.5\text{-}\mu\text{m}$ Mylar foil (contributing a minimal attenuation to impinging alpha particles) and stored in a light-tight box for alpha dosimetry purpose. Evidently, the $2.5\text{-}\mu\text{m}$ (thin) Mylar foil becomes extremely fragile during handling. Hence, utmost precautions should be taken while cutting the foil using a sharp scalpel. The second section (area = 25 cm^2) was used for thickness estimation of the dye layer. This film section was weighed using a chemical microbalance (PS-H200; Voltcraft, Taiwan, China) with an accuracy of ± 0.1 mg. The dye layer was removed using warm (approximately 40°C) distilled water, and the remaining polyester substrate was dried thoroughly in a hot air cabinet and weighed again. The dye layer thickness (t_D) was estimated as follows:

$$t_D = [(m_F - m_P)/F]/\rho_D \quad (1)$$

where, m_P , m_F , F and ρ_D are the mass of the film section (0.517 g), polyester substrate only (0.459 g), film area (25 cm^2) and density of the tissue equivalent dye material (1 g cm^{-3}), respectively. By substituting the numerical values in Equation (1), the thickness of the dye layer (t_D) was calculated to be $23.2 \text{ }\mu\text{m}$,

17% lower than the manufacturer's quoted value $28 \text{ }\mu\text{m}$. For dye layer thickness estimation, the same radiochromic film section (area = 25 cm^2) was weighed twice, with the dye layer and layer removed. Therefore, the uncertainty in the weight estimation will be the major contributor in error calculation of dye layer thickness than the uncertainty in area measurement.

Alpha spectrometry of the ^{241}Am source

We have used a thin, electrodeposited⁹ alpha reference source (activity: 560 Bq ; spot diameter: 7 mm , serial number: 64–98 RA; Amersham International, Amersham, UK) for irradiation purpose. The alpha spectra of the bare source and covered with $2.5\text{-}\mu\text{m}$ Mylar foil were recorded using a high-resolution Si(Li) surface barrier detector (LEC 200-2000, active thickness: $200 \text{ }\mu\text{m}$ mounting diameter 28.50 mm ; Canberra Industries, Inc., Meriden, CT) interfaced to a 1459 channel computerized analyser. The high-voltage supply, signal gain and detection time (live) were 80 V , 2048 and 605.9 s , respectively. The measurements were carried out in a dedicated alpha spectroscopy vacuum chamber. The alpha spectra are depicted in Figure 2.

Alpha exposure and evaluation of radiochromic films For the alpha exposure, the authors have prepared four pieces of $2 \times 3\text{-cm}$ samples cut from the modified EBT3 film sheet (area = 25 cm^2), whereas the sensitive (dye) surface was covered with $2.5\text{-}\mu\text{m}$ Mylar foil. The alpha source was placed at contact with the film and sequentially exposed for 5 , 10 , 24 and 94 h . After completion of alpha exposure, each film sample was cut in two pieces, the upper exposed part and the lower unexposed part (control) as depicted in Figure 3.

As the result of alpha exposure, the films turned “bluish dark”; furthermore, this colouration effect is estimated in terms of net optical density (NOD) defined¹⁰ as the difference between the optical densities of exposed film (OD_{exp}) and that of the control film (OD_{cont}):

$$\text{NOD} = \text{OD}_{\text{exp}} - \text{OD}_{\text{cont}} \quad (2a)$$

or

$$\text{NOD} = \log_{10}(I_0/I_{\text{exp}}) - \log_{10}(I_0/I_{\text{cont}}) \quad (2b)$$

where, I_0 , I_{exp} and I_{cont} stand for intensities of incident light and transmitted light through exposed and control (unexposed) films, respectively. The authors have used a simple handheld spot densitometer (DensiX; PTW Freiburg, Freiburg, Germany)

Table 1. Showing the fitting polynomials relevant to functions: alpha range and energy in the Mylar and EBT3 dye material (tissue equivalent) as depicted in Figure 5

Function	Stopping material	
	Mylar	EBT3 dye (tissue equivalent)
Range = $f(\text{Energy})$	(1) $y = 6.11 \times 10^{-01}x^2 + 2.64 \times 10^{+00}x + 5.90 \times 10^{-01}$, $R^2 = 1.00 \times 10^{+00}$	(3) $y = 7.43 \times 10^{-01}x^2 + 3.18 \times 10^{+00}x + 3.71 \times 10^{-01}$, $R^2 = 1.00 \times 10^{+00}$
Energy = $f(\text{Range})$	(2) $y = 4.55 \times 10^{-05}x^3 - 4.92 \times 10^{-03}x^2 + 2.79 \times 10^{-01}x - 5.50 \times 10^{-02}$, $R^2 = 1.00 \times 10^{+00}$	(4) $y = 3.05 \times 10^{-05}x^3 - 3.71 \times 10^{-03}x^2 + 2.35 \times 10^{-01}x - 1.23 \times 10^{-03}$, $R^2 = 1.00 \times 10^{+00}$

Table 2. Presenting the alpha exposure time, time-integrated specific surface activity (A), net optical density (NOD) of the film samples (Figure 3) and the corresponding alpha (D_a) and background gamma¹³ (D_g) doses

Sample ID	Exposure time (h)	A (kBq cm ⁻² h)	NOD	D_a (Gy)	D_g (Gy)
–	0	0	0	0	0
(a)	5	7.3	0.04	4.9	1.15×10^{-6}
(b)	10	14.6	0.07	9.8	2.30×10^{-6}
(c)	24	35.0	0.15	23.4	5.52×10^{-6}
(d)	94	137.2	0.75	91.6	2.16×10^{-5}

for the explicit estimation of the optical densities of the exposed (OD_{exp}) and control (OD_{cont}) films.

The time-integrated specific surface activity (A) was calculated as:

$$A \text{ (kBq cm}^{-2} \text{ h)} = Qt / \pi r^2 \quad (3)$$

where Q (0.56 kBq), r (0.35 cm) and t (h) are the alpha source activity, source radius and exposure time, respectively. By substituting the numerical values in Equation (3), the value of A was calculated to be $1.45t$ (kBq cm⁻² h).

Alpha energy loss and depth-dose calculation

The optical densities of the exposed films are evidently directly proportional to energy loss (i.e. energy dose) of the impinging alpha particles¹¹ reaching the film layer after passing through the 2.5- μ m Mylar foil and specific surface activity of the alpha source (Figure 4). The alpha energy and range calculations in the Mylar and tissue equivalent dye layer⁷ of the film material were performed using the range/energy and energy/range functions (Figure 5) constructed using the reference data.¹² The fitting polynomials relevant to these functions are presented in Table 1. The rest energy of the alpha particles (initial energy: E_0) traversing 2.5- μ m Mylar foil (E_1), energy loss within 23- μ m thick dye layer (ΔE) and the exit energy (E_2) were calculated using the fitting polynomials (Table 1) as presented below:

- Step 1: Set initial alpha energy E_0 and calculate the range in Mylar (R_M) using polynomial (1), $R_M = f(E_0)$.
 Step 2: Calculate the range difference ($R_M - T_M$) and exit alpha energy (E_1) using polynomial (2), $E_1 = f(R_M - T_M)$.
 Step 3: Calculate the range (R_D) of alpha particle of energy E_1 in the dye layer using polynomial (3), $R_D = f(E_1)$.
 Step 4: Calculate the range difference ($R_D - T_D$) and the corresponding energy difference (ΔE) in the dye layer using polynomial (4), $\Delta E = f(R_D - T_D)$.

Step 5: Calculate the exit energy (E_2) from dye layer as flows: $E_2 = E_1 - \Delta E$.

Step 6: The remaining energy E_2 is completely lost in the sufficiently thick polyester layer ($T_P = 100 \mu\text{m}$) as shown in Figure 4.

Alpha dose delivered in the energy loss (ΔE) volume (v) in radiosensitive dye is given as:

$$D_a \text{ (Gy)} = \Delta EAS / \Delta M \text{ (J kg}^{-1} \text{)} \quad (4)$$

where alpha energy loss ΔE (in mega electronvolts) expressed as ($\Delta E / 6.24 \times 10^{12}$) (in joules) = $1.60 \times 10^{-13} \Delta E$ J; time-integrated specific alpha surface activity A (in kBq cm⁻² h) expressed as (3600×1000) (in Bq cm⁻² s) = $3.6 \times 10^6 A$ Bq cm⁻² s; the surface area S (Figure 4) was calculated to be $\pi \times (0.7/2)^2 \text{ cm}^2 = 0.385 \text{ cm}^2$; alpha energy loss volume (v) considered to be a 23- μ m long 0.7-cm diameter right cylinder (Figure 4), $v = 23 \times 10^{-4} \pi \times (0.7/2)^2 \text{ cm}^3 = 8.85 \times 10^{-4} \text{ cm}^3$; consequently, the mass (ΔM) of stopping (dye) material ($\rho = 1 \text{ g cm}^{-3}$) within the energy loss volume calculated as: $8.85 \times 10^{-4} \text{ cm}^3 \times 1 \text{ (g cm}^{-3}) = 8.85 \times 10^{-4} \text{ g} = 8.85 \times 10^{-7} \text{ kg}$.

By substituting the numerical values in Equation (4), we get:

$$D_a \text{ (Gy)} = (1.60 \times 10^{-13} \times 3.6 \times 10^6 \times 0.385 \times 1.13 \times 10^6) \Delta EA$$

or

$$D_a \text{ (Gy)} = 0.251 \Delta EA \quad (5)$$

Gamma background estimation

Alpha radionuclides, including ²⁴¹Am also emits γ -rays.¹³ The authors have placed a Geiger-Mueller (GM) counter-based

Table 3. Showing important alpha emitters¹⁴ with corresponding half-lives and energies (E_0), calculated entrance (E_1) and exit (E_2) energies and energy loss (ΔE) in the sensitive layer (Figure 5) and the dose normalization factor (k_a) relative to ²⁴¹Am

Alpha emitter	Half-life	E_0 (MeV)	E_1 (MeV)	E_2 (MeV)	ΔE (MeV)	k_a
²¹⁰ Bi	5.01 days	4.67	4.40	3.17	1.24	2.15
²⁴¹ Am	450 years	5.48	5.21	2.55	2.66	1.0
²²³ Ra	11.4 days	5.70	5.43	2.42	3.01	0.88
²²⁵ Ac	10 days	5.94	5.67	2.29	3.37	0.79

Figure 6. (a) The calibration graph representing the time-integrated specific surface activity (A) of alpha particles from the 560-Bq ^{241}Am reference source as a function (inset) of net optical density (NOD) of the modified EBT3 radiochromic film. (b) Calibration graphs of alpha emitters ^{210}Bi (1), ^{223}Ra (3) and ^{225}Ac (4) evaluated using the experimentally estimated energy loss normalization factor k_a (Table 2) and calibration function of ^{241}Am reference (2).

Gamma-probe (FHZ 632; Thermo Electron GmbH, Erlangen, Germany) interfaced to a Gamma-monitor (FH 40G-L10; Thermo Electron GmbH) at contact with the ^{241}Am reference source. The 0.2-mm-thick hard-aluminium entrance wall of the GM probe eliminates the alpha particles totally, thereby enabling the assessment of unperturbed gamma dose rate.

RESULTS

The values of NOD, time-integrated specific alpha surface activity (A) of ^{241}Am reference source and corresponding alpha dose (D_a) were calculated using Equations 2a, 3 and 5, respectively, and are presented in Table 2.

The energies of alpha emitters of interest,¹⁴ ^{210}Bi , ^{241}Am , ^{223}Ra and ^{225}Ac at the entrance (E_1) and exit (E_2) points of the radio-sensitive EBT3 dye layer (Figure 4) and related energy loss (ΔE) were evaluated using calculation Steps 1–6 as described in “Alpha energy loss and depth-dose calculation” section. Furthermore, the alpha energy losses were normalized against that of ^{241}Am reference, and the normalization factors (k_a) are presented in Table 3.

Time-integrated specific alpha surface activity (A) of the ^{241}Am reference source was plotted as a linear function (inset) of NOD, thereby yielding the calibration graph (Figure 6a). Evidently, the NOD is directly proportional to alpha dose (D_a). On the other hand, D_a depends on both alpha energy loss (ΔE) and time-integrated specific surface activity (A) as described in Equation (5). Hence, NOD could be expressed as an explicit function of ΔE and A . Hence, we have used the alpha energy loss normalization factors (k_a) to parameterize the calibration graph for ^{241}Am

reference source in order to construct the calibration graphs for other alpha emitters of interest (Figure 6b).

The unperturbed gamma dose rate at contact with the ^{241}Am reference source was estimated to be $2.3 \mu\text{Gy h}^{-1}$. The calculated gamma background doses are presented in Table 2.

DISCUSSION

The authors have developed a novel alpha dosimetry technique based on radiochromic films widely used in photon dosimetry and a low activity, thin ^{241}Am reference source. Most of the alpha radionuclides also emit low-intensity γ -rays, and the conventional QA protocols validate those γ -rays using sophisticated gamma spectrometers. The present method is based on cost-effective radiochromic films and a user-friendly handheld optical densitometer. Furthermore, the use of 2.5- μm (thin) Mylar foil wrapping the radiosensitive layer of the film enabled the detection of low-energy alpha particles down to approximately 4 MeV.

This technique, however, suffers from three major shortcomings. (a) Owing to longer exposure time (Table 2), it is suitable for assessment alpha emitters of relatively longer half-lives, *i.e.* in the order of several days (Table 3). (b) During routine applications, thick alpha sources *i.e.* alpha radiopharmaceuticals of interest absorbed in small pieces of filter paper will be deployed. This will cause a substantial “self-absorption”, resulting in reduction of specific surface activity (Equation (3)) and energy loss of the alpha particles. To overcome this limitation, an alpha self-absorption correction factor¹⁵ will be taken into account. (c) Loss of dye material (sensitive layer) while removing the protecting polyester sheath (Figure 1). Hence, the sensitive layer thickness of the modified EBT3 radiochromic film sheet should be estimated using the method described in this article prior to dosimetric applications.

Majority of alpha-emitting radionuclides also emit γ -rays and β particles, however, of very low intensities.¹³ In this work, the gamma (background) dose contribution was estimated to be negligible in comparison with alpha dose (Table 2).

The authors already have developed a radiochromic film-based alpha dosimeter, considering all radiological safety aspects,¹³ in particular, contamination hazard. Application of this user-friendly method for activity calibration and QA procedures of radiopharmaceuticals incorporating alpha emitter is envisaged. Those drugs are the principal candidates of highly efficient TIT for the treatment of micrometastatic ailments.¹ Scientific collaboration with a leading teaching hospital in Sydney, Australia, involved in clinical trials of ^{223}Ra -dichloride (Alphadrin) for the treatment of bone metastasis of prostate cancer patients is in progress.

ACKNOWLEDGMENTS

The authors would like to thank Mr Marek Dolleiser of School of Physics, The University of Sydney, Australia, for his professional support during alpha spectrometry experiments. Assistance of Mr Wolfgang Deya of Institute of Nuclear Medicine, University Hospital Essen, Germany, is also thankfully acknowledged.

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