RADIOCHROMIC FILM DOSIMETRY

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Introduction: Chemical dosimeters are based on the concept of a radiation sensitive chemical reaction of which the amount of reaction products is correlated with the absorbed dose to their active elements. Among them, coloration dosimeters have a rich and interesting history in dosimetry. For example, polyacetylenes (PDAs) form a unique class of polymeric materials that upon irradiation join-up aligned and their conjugated backbones and grow in length with the level of exposure. The thus formed polymeric chains are responsible for the strong optical absorption, turning blue or red depending on the specific composition each PDA type. The degree of material coloring depends on the energy imparted to the color producing elements (chromophores) and the space between the last monomer unit in the polymeric chain and the adjacent monomeric unit. The optical absorption can be modified in the majority of PDAs by various stimuli, such as exposure to heat (thermochromism), light (photochromism) and application of mechanical stress (mechanochromism).

Polyacetylenes can be structured in various forms, such as bulk materials, multilayer and monolayer films, and even incorporated into organic host matrices to form nanocomposites. For example, radiochromic (RC) films are self-developing coloration passive detectors (no chemical processing is required, as opposed to silver halide films, however they need some time for full development) consisting of one or more radiation sensitive-layer of diacetylene microcrystals on a thin organic base. Under irradiation of the initially almost colourless monocrystals, chromophores become coloured through a chemical reaction due to the energy imparted to them (Fig.1). In principle, but not always in practice, the degree of colour formation is proportional to the energy imparted to them.

Colourization is usually quantified as the ratio of intensities of the transmitted (I_t) to the incident (I_o) light of a specific spectrum to an object, or by its optical density (OD), defined as $\log_{10}(I_o/I_t)$. For example, in case of OD=1.00, 10% of the incident light is transmitted. Moreover, the netOD, the difference between the OD of an irradiated and an unirradiated similar object ($\Delta O=\log_{10}(I_{t,unirad}/I_{t,irr})$) is also often used.

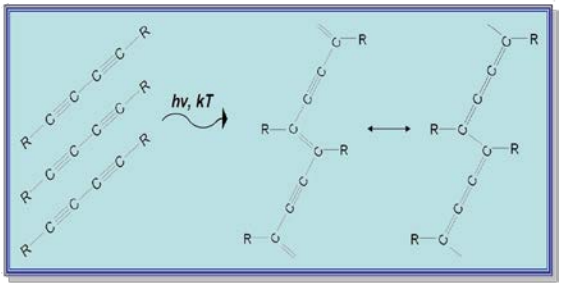

Fig. 1 Disubstituted diacetylenes, such as pentacosa-10,12-diyonic acid, are polymerized following exposure to ionizing radiation (ISP, Wayne, NJ, USA).

Fields of use: High spatial resolution, weak energy dependence, the easy use make RC media suitable for mapping of radiation fields with even high dose gradients. Commercially available diacetylene microcrystals with high radiation reactivity and shelf-life are widely used, e.g. for:

- industrial radiography and radiation processing for the assessment of the absorbed dose and the 2D dose mapping, such as in medical device sterilization, food sterilization, mutation of seeds, insect pest control, mutation breeding of seeds, modification of mechanical and electrical properties of some materials,
- beam alignment and profiling, dose measurements in fields produced by low energy X-ray sources, linear accelerators, synchrotron and hadron accelerators, etc,
- nuclear reactor dosimetry,mapping and measurement of hot particles,
- security and military dosimetry, and
- triage after radiological accidents / incidences.

However, the prevalent field of use of the radiochromic media is the biomedical one, such as in a) radiation teletherapy (e.g. machine output, depth-dose measurements, dose profile and in general quality control of beams in conventional, stereotactic, IMRT microbeam and hadron radiation therapy, interface dosimetry and *in vivo* 2D dosimetry), b) brachytherapy (e.g. dose mapping in interstitial, intracavity and endovascular brachytherapy), c) blood irradiations, and d) radiology (e.g. QA and commission of radiological units, skin dose mapping in computed tomography and fluoroscopically guided interventions).

Various issues of RC films and their use has been reviewed in depth, such as by the American Association of Physicists in Medicine (Niroomad-Rad *et al*1998), Soares (2007), Williams& Metcalfe (2011) and by Devic (2011). However, the intent of this report is to present material from a user-oriented and practical standpoint.

Mechanism: Nowadays, the most widely used radiochromic molecules are disubstituted diacetylenes (R-C≡C-C≡C-R', where R and R' are monovalent organic substitutes, Fig. 1) present in GafChromic^{TM} films by International Speciality Products- Ashland (ISP, Wayne, NJ, USA). The corresponding molecules undergo a dose-dependent polymerization reaction in the blue polydiacetylene forminitiated among other things by exposure to ionizing radiation, provided that the monomers are packed in such a way that the triple bonds of the adjacent molecules. The colored crystals are built-up by packing thousands of monomer molecules into 3-D regular lattices along a direction fixed by the crystal lattice. Taking into account that the packing of the monomers in the microcrystals depends in large to on the particular end-groups, the radiation sensitivity depends among other things on the particular end-groups.

The colorless monomer pentacosa-10,12-diyonic acid (PCDA),usually in the form monomeric roughly spherical micro-crystals about 0.75 μm in diameter dispersed in a gelatin matrix, is the sensitive element of HD-810 (Fig.2), HDv2, MD55-2 (or MD-V255, Fig. 3), MD-V3, HS and XR-R films, which exhibit main absorption peaks, λ_{max}, at about 615 and 670 nm (Fig.4) at

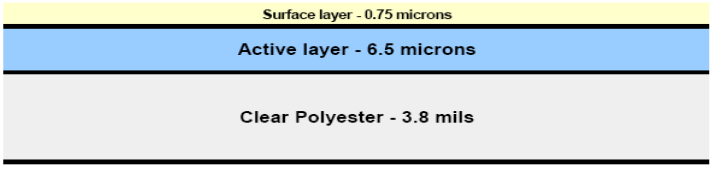

Fig. 2The structure of HD-810 films (a ~7μm PCDA active layer coated on a transparent polyester base and protected by a thin gelatine layer) often used in ~10 to 500Gy photon fields (HD-810 films were recently replaced by H-DV2-films with a ~8μm thick layer).

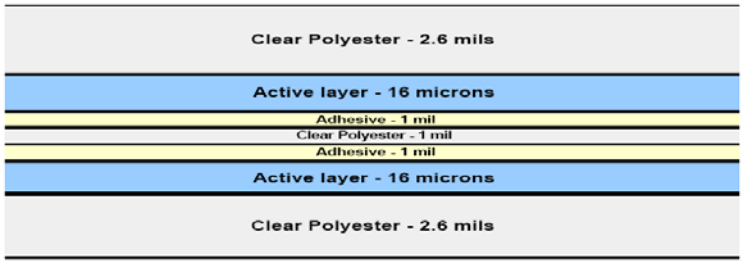

Fig. 3 The structure of MD-22 films (~two 16 μm PCDA active layers)often used in ~2 to 50 Gy photon fields (1 mils= 25.41 μm- ISP, Wayne, NJ, USA).

room temperature and a relative complicated dependence of the position of these maximum on dose and thermal history. For example, the almost transparent HD-810 and MD55-2 films are becoming blue after irradiation, due to the strong absorption of light in the red region. Similarly, lithium pentacosa-10,12-diyoniate (LiPCD) microcrystals, usually of larger size (often 10 to 50 -μm long hair-elements aligned more or less parallel to the coating direction of the film), are used in the more sensitive classes of the films, EBT (Fig. 5), MD V3, RTQA, XR-CT and XRQA also manufactured by ISP. LiPCD exhibits main absorption peaks at ~583 and 635 nm (Fig.6), with decreasing λ_{max} as the temperature increases from ~20 to 40 ^{o}C (~0.4 nm per 1 ^{o}C change, according to Rink *et al* 2008). Both PCDA and LiPCD undergo polymerization, provided that the carbon-carbon triple bonds of the adjacent molecules are at distance less than 1 μm (Fig.1).

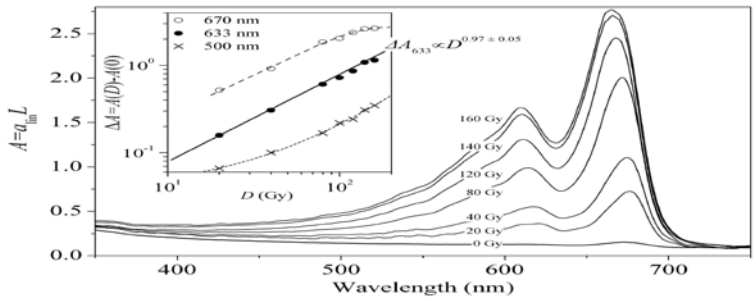

Fig. 4 Absorption spectrum of HD-810 films as a function of absorbed dose following ^{60}Co γ-irradiation at room temperature (the optical density, OD, of a film is the product of linear absorbance, A , with loge). Note the gradual shift of the peaks towards lower light wavelengths with increasing local polymer concentration related to increased dose, the low absorbance in blue region, as well as the practically linear increase of the net absorbance with dose at 633 nm in the studied range of doses (Koulouklidis *et al* 2013)

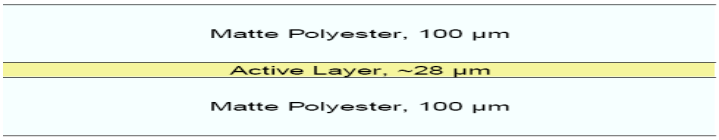

Fig. 5 The structure of EBT3 films (~30μm-thick LiPCD active layer (Z_{eff}=7.26,1.2 g/cm^3) Laminated between two matte polyester layers (Z_{eff}=6.64, 1.35 g/cm^3) assumed to have an energy independent response over the 0.1 to few MeV photon energy (the actual thicknesses may vary slightly). These films are currently widely used in teletherapy, mainly for both QA and skin dosimetry.

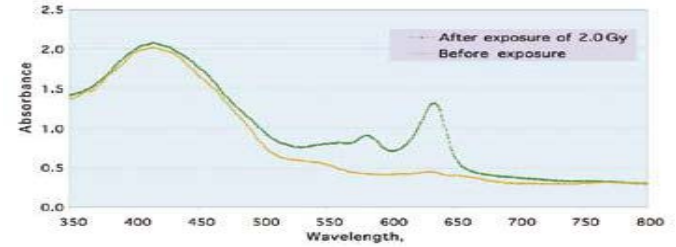

Fig. 6 Absorption spectrum of an EBT3 film following a 2.0Gy photon irradiation. Note the high sensitivity of LiPCDA loaded films *vs* those loaded with PCDA, the differences in the peak wavelengths, λ_{max}, the stability of λ_{max} with dose and the large absorption light of the unexposed films in the region of small wavelengths, resulting to the yellow appearance of the unirradiated films (ISP, Wayne, NJ, USA).

Characteristics: Among the most important characteristics of the 2D dosimeters are:

- dynamic range of measured doses that has to fit the needs of the study,
- energy invariant response of the dosimeters to the type of radiation to be studied,
- response independent of dose rate and dose rate fractionation,
- small or even no angular dependence of their response,
- temporal stability of the readings coupled with minimum influence of environmental conditions on sensitivity before irradiation, pre- and post- irradiation and during reading (e.g. temperature, pressure, humidity, water resistance, and UV light resistance, such as that that to fluorescent light and sunlight),
- spatial resolution and response uniformity adequate for the needs of the application,
- minimum perturbation of the irradiation field due to the presence of the dosimeter (e.g. small size, equivalence to the material where the films are inserted),
- luck of light polarization effects and interference patterns during read-out,
- minimum batch to batch variations in response, and
- low cost, ruggedness and ease to use.

Energy response: The quantity often desired in medical applications is the absorbed dose to human tissues, such as in soft-tissue (water is often considered to be an appropriate soft-tissue substitute material for photon fields with energy higher than ~100 keV). The energy dependence of RC film response relies on the specific composition of the various layers of each film type, which has been shown to vary between batches in some cases. The ratio of the mass- absorption-coefficients of both PCDA and LiPCD to water is almost constant in case

of photons of energy higher than ~150 keV, dropping to values lower than 1.0 at lower energies, but higher than 0.5, due to the reduced Z_{eff} for photoelectric effect in the active elements relative to water. Therefore, some manufactures added some materials of higher Z_{eff} in the surrounding layers in an attempt to compensate for the underesponse to low energy photons. Therefore, unlike radiographic films where the active materials are some silver halides and therefore, have a high Z_{eff} for photoelectric effect, RC films usually made of low Z organic active materials are not only closer to soft tissue equivalence in photon fields but also exhibit, at least in general, a response with very small angular dependence.

The GafChromic films with pentacosa-10,12-diyonic acid microcrystals as active monomers, other than thee of the XR familly that contain high Z materials to enhance their response to low energy photons, are often considered to be almost tissue equivalent to photons of energy higher than about 150 keV. At lower energies, e.g. Muench *et al* (1991) showed that the response HD810 films to 60 kVp x-rays (28 keV_{eff}) is lower by about 30% than that to 4 MV x-rays. Kron *et al* (1998) reported that MD-55 films underestimated the dose by a factor of two, when irradiated with 26 keV x-rays. Nariyama *et al* (2005) reported an almost constant under-response by 20% in HD-810 films in the energy region 30 to 100 keV relative to ^{60}Co gamma rays, and a gradual increase in MD55-2 film under-response from about 5% to almost 40% as the energy decreases in this energy region. Similarly, Cheung *et al* (2004) studying MD55-2 and HS film observed a gradual decrease with decreasing energy from 100 to 30 keV up to ~40% and a large over-response (up to a factor of five at 50 kV_{eff}), as anticipated in XR-T RC films. On the other hand, Brown *et al* (2012) reported responses to 35 keV photons of EBT, EBT2 and EBT3 films that are loaded LiPCD microcrystals of 0.76, 1.24 and 0.98 relative to 4 MV X-rays, respectively.

Post-irradiationcolor stability: Taking into account that polymerization proceeds for some time after irradiation, development continues even after the termination of the irradiation (Fig. 7), thus introducing some dose-rate and dose fractionation effects in real-time dosimetry.

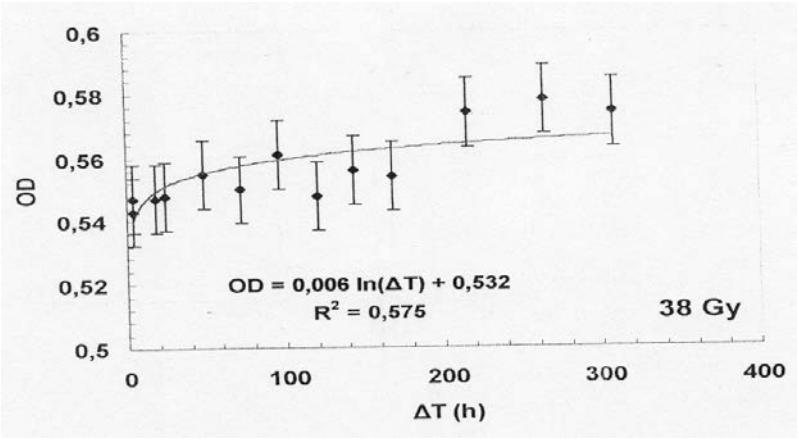

Fig. 7 Optical density (OD) vs the elapsed time (ΔT) after the irradiation of MD55-2 film at 38 Gy ^{60}Co γ-irradiation and read in the red channel with a flatbed document scanner (Bazioglou & Kalef-Ezra, 2001).

Environmental effects: Molecular motions influence the polymer structure. Therefore, the shape of the absorption spectrum is influenced by the temperatures during film irradiation, storage and reading, usually shifting towards lower wavelength with increasing reading temperature (Fig. 8), a factor which has a substantial impact on measurements carried out using a very narrow wavelength light-band. Moreover the spectral shape is dose-dependent in films loaded with PCDA, and to lower extent in those with LiCPD. In addition, humidity and ultraviolet exposure (even by sunlight or light from florescent lamps depending on the coatings) may influence the film response by a degree which depends among other things, on the coatings used. Taking into accounts that RC films undergo post-exposure signal intensification, with the polymerization rate decreasing with time, adequate time has to be elapsed between irradiation and measurement to achieve accurate measurements. For example, readings of LiPCD and PCDA containing films are often carried out with some delay, such as 1 and 3 days post-irradiation, respectively, assuming thereafter a stable and dose-rate insensitive response. Therefore, sticking to a fixed carefully designed protocol, as any other dosimetric method, is crucial to obtain reproducible and accurate dosimetric results with RC films.

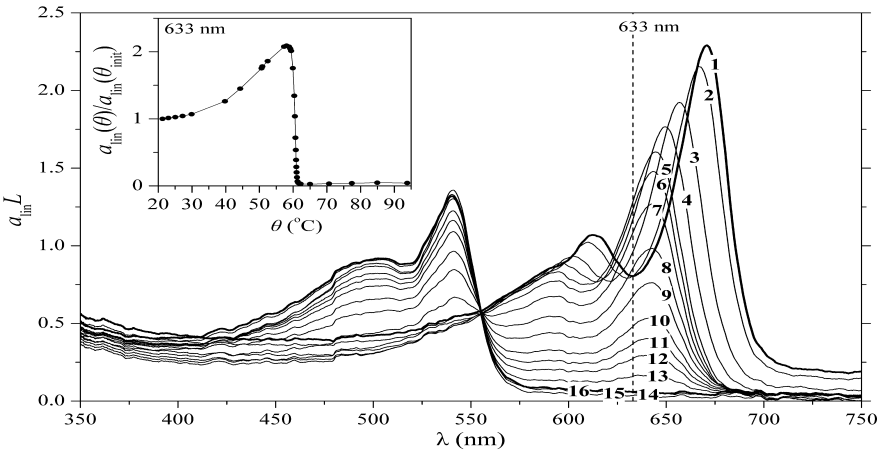

Fig.8. Linear absorbance, $A=a_{lin}\cdot L$, spectra as a function of reading temperature, θ_{read} in HD810 films given 100 Gy of ^{60}Co γ-rays (θ_{read} (°C)= 21.3 (1), 29.8 (2), 44.3 (3), 4: 52.5 (4), 58.0 (5), 59.4 (6), 59.9 (7), 60.3 (8), 60.5 (9), 60.7 (10), 60.8 (11), 60.9 (12), 61.0 (13), 65.0 (14), 85.0 (15), 93.8 (16)). As the temperature rises up to ~58 °C the blue form maxima are continuously shifted towards shorter wavelengths. A clear thermochromic transition from the blue phase to the red phase takes place at higher temperatures. The inset shows the the fractional absorbance at temperature θ with respect to its initial value (θ_{init}) for λ=633 nm (Koulouklidis *et al*, 2013).

Light sources and readers: Dose assessments are often carried out using densitometers, i.e. devices which measure light transmission through a sample, such as scanning densitometers that allows for transmission measurements at many points over a sample surface in an automated fashion with adequate spatial resolution for the study.The interaction of light with an RC film depends on the absorption and scattering at the various film layers. Light sources of

appreciable red spectral content, such as He/Ne laser (633 nm), diode lasers (650-670 nm) and wideband red-emitted LED sources coupled with band-filters are often used along with either a photodiode or a photomultiplier allowing for point by point analysis Alternatively, Xe cold-cathode ray tubes or "white" light sources are used, such as those used in spectrophotometers and in flatbed color document scanners equipped with "white" light sources and arrays of charged coupled devices (CCD) similar to those used in digital optical cameras. In the latter case, the transmission (or the reflection) images are analyzed for dose assessment in three wide color channels (RGB analysis, Fig.9)) and the data obtained in either one of them (usually in the red one) or in all channels (multichannel reading - Kalef-Ezra & Karava 2008, Micke *et al* 2011) and adequate signal resolution, such as a 12 bit signals that provides 4096 shades of gray. For example the OD of MD52-2 films assessed using the R channel following irradiations at various doses with ^{60}Co and 6 MV X-ray beams at Ioannina University Hospital (IUH) is shown in Figs. 10,11 after the required time for the warm-up of the scanner lamp (Fig.12), as well the ΔOD increase in case of irradiation of EBT3 films as at the three light channels (Fig.13). Note the difference in sensitivity between EBT3 and MD-55 films by about an order of magnitude (typical EBT3 dynamic range ~20 mGy to 20 Gy).

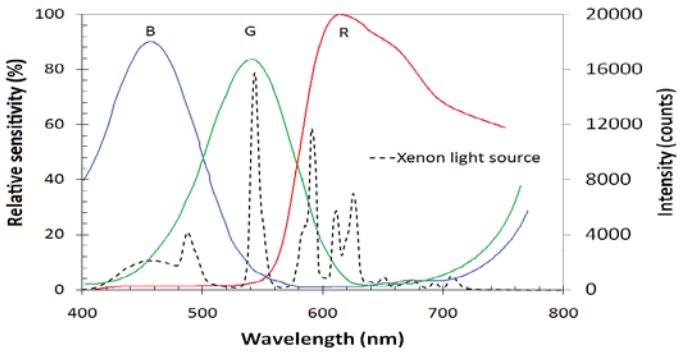

Fig.9 Spectral sensitivities of the red, green and blue channels of a CCD of the type used in flatbed scanners, also shown is the emission spectrum of the Xenon cold cathode ray tube used in the EPSON 10000XL flatbed scanner (Williams & Metcalfe, 2011).

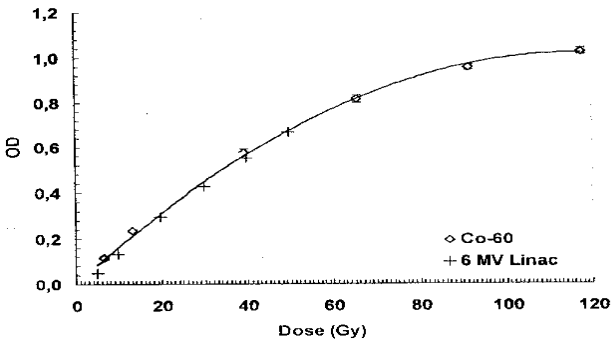

Fig 10 Optical density (OD) of MD55-2 films4 days post-irradiation (red component) using a VM355 Infinity document scanner (Bazioglou & Kalef-Ezra, 2001).

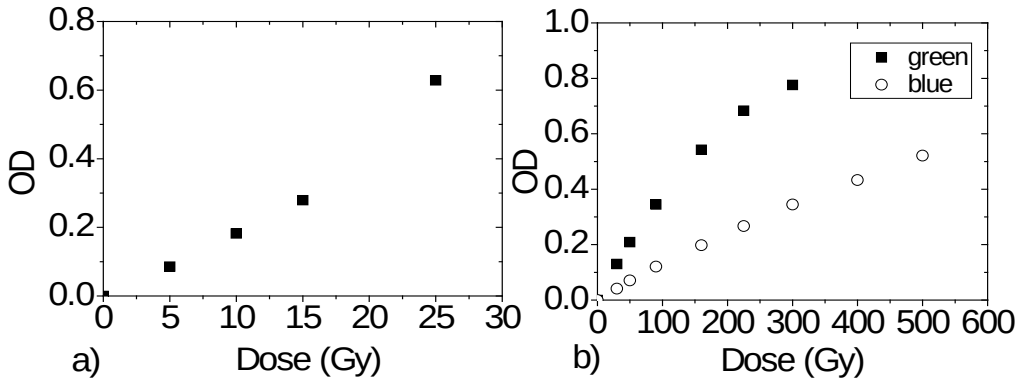

Fig.11 Optical density of MD55-2 assessed 4 d post-irradiation with ^{60}Co γ-rays using RGB analysis with a document scanner. Note the use of the non-red component allows the extension of the dynamic range to the 1 kGy region (Kalef-Ezra & Karava, 2008).

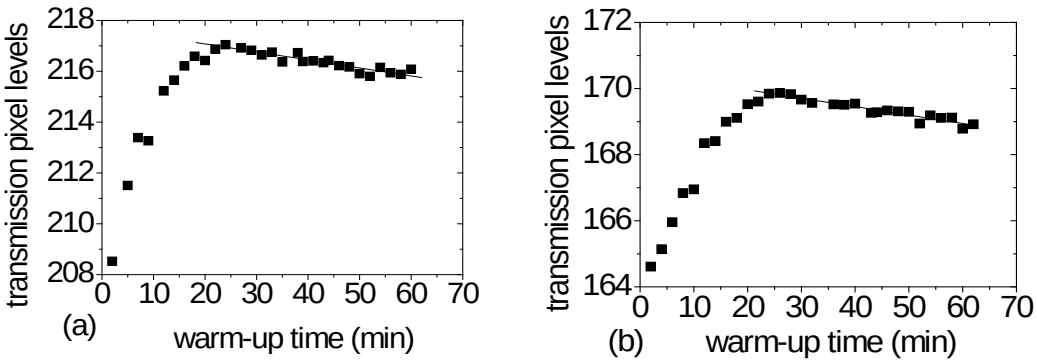

Fig. 12 Scanner warm-up effects by multiple readings at the R channel ofMD5-2 films (a) non-irradiated and (b) irradiated with 5 Gy of ^{60}Co γ-rays (b) four days before reading (Kalef-Ezra & Karava, 2008).

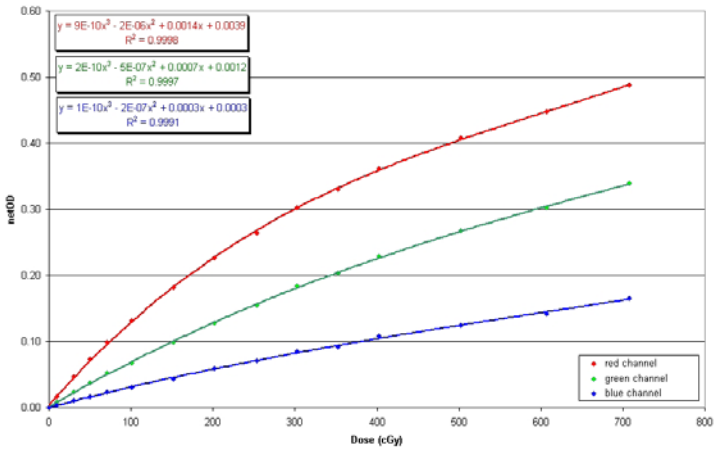

Fig.13 The net optical density, netOD, of EBT3 films irradiated with high energy photons assessed with a flat document scanner (RGB analysis) versus absorbed dose in water.

The film transparency can be assessed alternatively by measuring the intensity of the reflected light (double transmission plus reflection) rather the one of the transmitted one (Kalef-Ezra & Karava 2008). Moreover, in some film types, such as those of the XR family,

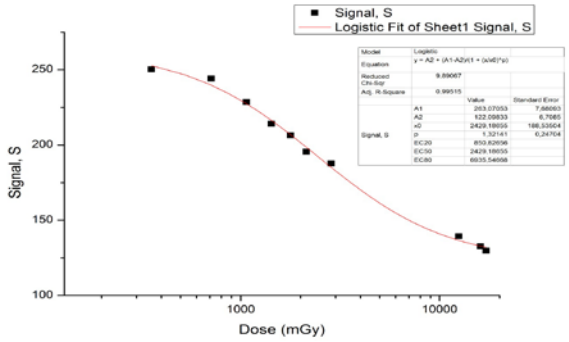

Fig.14 The reflection signal of XR RV3 films following irradiation with 662 keV γ rays (^{137}Cs) using the red component of the RGB image using a Scanmaker s480 document scanner (the 17 μm-thick active layer is laminated between two binding layers and two 97 μm- thick polyester layers, a yellow and a white one, resulting in almost full reflection of the light.

one of the non-active layers reflect equally all light wavelengths (white material), thus allowing only for reflection measurements (Fig. 14). In attempt for use of such films in the field of radiology, where the doses in water are lower than those in therapy, some high Z material is often added, such as barium salts to enhance their response to low energy photon. As anticipated the response of such films exhibit substantial energy response. For example, as shown in Fig.15 the maximum sensitivity of XR-RV2 films, defined as the dose in water required for radiation induced change of the measured signal by 50 AU, is achieved using a 100 kV_p X-rays spectrum. The response of these films (tailored for use in skin dose mapping following fluoroscopy guided interventions) decreases as the mean photon energy decreases below the maximum (an almost 3-fold reduction at 50 kV_p) as well as at higher photon energies (e.g. an almost 7-fold reduction at 6 MV X-ray fields).

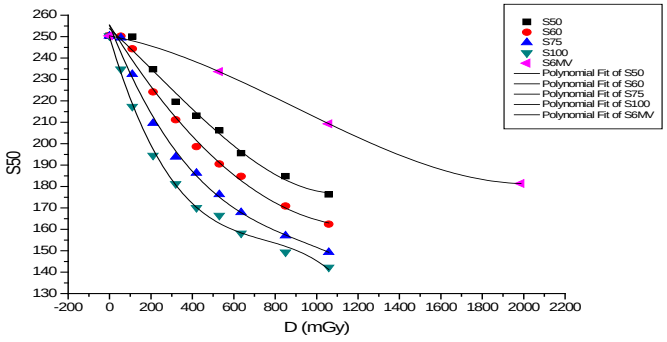

Fig.15 Calibration of XR-RV2films to various photon fields measured with an HP Scanjet 4570c document scanner in the reflection mode. Note the large energy dependence of this type of film due the barium salts to enhance their response to low energy photon widely used in fluoroscopically guided interventions for skin dose mapping (maximum sensitivity to ~100 kV_p X-rays- Kalef-Ezra *et al* 2010).

Spatial resolution: PCDA molecules are usually packed as flat grains about 2 μm in size, while LiPCD molecules as aligned 10 to 50 -μm long hair-elements. Based on the physical size of the crystals, PCDA containing films are anticipated to exhibit superior intrinsic spatial

resolution than those containing LiPCD. However, in most cases, the spatial resolution is governed by the resolution of the reader rather than the inherent resolution of the film itself.

Uniformity: A pre-request for 2D accurate dosimetry is the uniform response of the dosimeter over both very small and large distances from its center. In case of RC films it is often assessed by cutting films into small pieces, e.g. ~1cm x 1cm and irradiating them in the same field at different doses, in groups of at least six films per dose, a procedure that bypasses the potential problems that are related with spatial non-uniformities of the scanner. The coefficient of variation of readings (the ratio of the standard deviation to the mean value) of such pieces of a single HD-810 film after irradiation of the randomly chosen pieces at various doses at IUH and read using both scanning modes using the R channel of the RGB image is shown in Figs. 16,17.

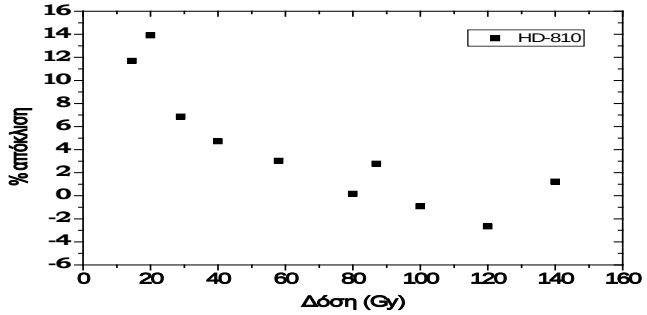

Fig. 16 The coefficient of variation of dose measurements of HD810 films irradiated at various doses of 1.25 MeV γ–rays using the R component obtained in the transmission mode

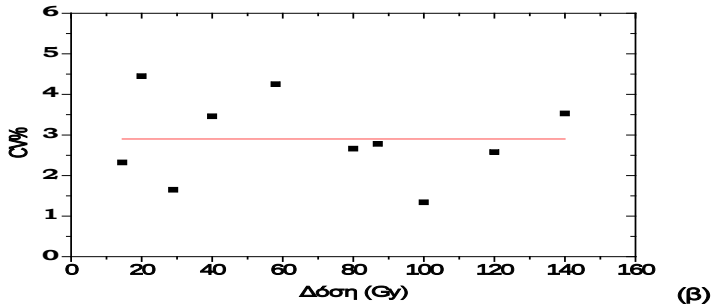

Fig. 17 The coefficient of variation of dose measurements of HD810 film previously irradiated at various doses of 1.25 MeV γ–rays using the R component obtained in the reflection mode.

High-LET fields: Radiochromic films can be also used in high LET fields. However, as many other dosimetric materials, the RC sensitivity (the change of their reading per Gy) is lower following high LET irradiations (e.g. alpha particles and other heavy ions, fast neutrons) than that following of low LET irradiations (β-particles, high energy electrons, γ- and X-rays). For example, EBT films were found by Reinhardt et al (2012) to under-respond up to 5% to proton energy below 40 MeV and up to 20% in the vicinity of the Bragg peak in case of 200 MeV proton beam. In addition, the signal produced to MD55-2 per Gy in soft tissue from fast neutrons of a particular spectrum was found to be ~46% of the signal per Gy in soft tissue from ^{60}Co γ-rays In addition, the irradiation of such films with a thermal neutron fluence of

10^{10} n_{th} cm^2 resulted in a signal similar to that following a 4 mGy irradiation with ^{60}Coγ-rays (Bazioglou & Kalef-Ezra, 2001).

Radiochromic 3D detectors: Nowadays, 3D radiochromic dosimeters are also commercially available. They usually consist of a clear polyurethane matrix doped with RC components generate a color change, and hence optical absorption or optical density (OD) changes, on exposure to ionizing radiation. PRESAGE® is such a plastic dosimeter based on transparent polyurethane mixed with a leucomalachite green dye and a number is organic and/or metallic indicators. Its irradiation results in local change in OD, usually assessed using as light either a He/Ne laser (633 nm) or a light-emitting diode that emit light of appropriate wavelength along with an appropriate camera, that allows to obtain CT images (Bräuer-Krisch *et al* 2015).

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