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CONCEPTUAL BASIS FOR THE RADIOCHROMIC DYE FILM DOSE METER AS A TEST OF PARTICLE TRACK THEORY

Johnny W. Hansen

Abstract. This report is a summary of a lecture held at the Danish-Polish Symposium on Radiation Chemistry in Warsaw, October 1979, describing an initiated work connected to the particle track theory worked out by R. Katz and coworkers. A short description is given of the theory and the applicability of the theory in the use of the radiochromic dye cyanide film dose meter as a detector in radiation of different qualities. A few experimental results are given.

INIS descriptors: COLORIMETRIC DOSEMETERS, CYANIDES, DOSE-RESPONSE RELATIONSHIPS, DYES, IONIZING RADIATIONS, LET, PARTICLE TRACKS, RADIATION DETECTORS.

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1. INTRODUCTION

In collaboration with Professor Robert Katz, Behlen Laboratory of Physics, University of Nebraska-Lincoln, USA, and Dr. Mikael Jensen, National Institute of Radiation Protection, Stockholm, Sweden, work has been initiated in connection with the particle track theory worked out by R. Katz and coworkers. Until now contributions to the work consist mainly of an experimental investigation of the applicability of the theory to the use of the radiochromic dye cyanide film dose meter as a detector in radiation of different qualities, i.e. low- and high-LET radiations.

The dye film dose meter has been investigated in our laboratory[1,2], and on the basis of some of these results the system appeared to be a suitable detector for testing the Katz particle track theory. Besides testing of the track theory these experiments can provide further knowledge of the dose meter response characteristics and might give insight to mechanisms of its response to ionizing radiation, namely that of dye formations from the leuco cyanide dye precursor.

2. CHARACTERISTICS OF THE FILM DOSE METER

The main features of the detector will briefly be summarized. The radiation detector used in these experiments is a thin-film plastic dose meter developed for measurements of high absorbed doses and dose distributions in intense radiation fields. The dose meter is commercially available from Far West Technology, 330 S. Kellogg, Goleta CA 93017, USA.

The host material of the dose meter is Nylon containing 10-15% of a colourless radiochromic dye precursor that becomes deep blue coloured upon irradiation, the intensity of colouration being proportional to the energy absorbed by the film. The dye precursor, which is used as the radiochromic sensitive component, is a hexa(hydroxyethyl)pararosaniline cyanide $[C_6H_4N(C_2H_4OH)_2]_3C\text{-}CN$ dissolved in Nylon $(C_{12}H_{22}N_2O_2)_n$ as the matrix material. Upon irradiation the C-CN bond is hetero genously broken and the triarylmethane groups become the highly coloured carbonium ion. This reaction takes place upon irradiation with particle energies exceeding the C-CN bond strength, which is about 3.8 eV.

The formed dye has a broad optical absorption band in a part of the visible spectrum having a maximum at 601 nm. Optical spectra for both irradiated and unirradiated film are shown in Fig. 1. The measured optical density increases linearly with dose over a wide range, Fig. 2. For the thin-film ($5\ mg/cm^2$) dose meter, which we use, the linearity covers a useful absorbed dose range of $5 \cdot 10^4$ to 10^7 rad[(3)], and there is in this dose interval no apparent effect of change in dose rate in the range from 1 to 10^{14} rad/s[(4)]. Investigations have also shown no energy dependence of the radiation response characteristic of the dose meter over a wide range of photon and electron energies (0.01 - 10 MeV)[(5)].

In these experiments the tested endpoint, the blackness, is expressed as increase of optical density per unit film thickness (ΔOD/mm) and is measured at the wavelength of 510 nm, which is off the maximum at the edge of the absorption band, where the film shows less sensitivity to radiation. This is due to too high an optical density at 601 nm for measurement, which causes the spectrophotometer to saturate before the film optical density shows saturation. For radiation energies which we have used in previous studies, the linear energy transfer in the dosimetric material is less than 4 MeV/cm. For these radiations, the dose meter response to equal absorbed doses has by previous experiments been found to be constant for absorbed dose rates up to 10^{14} rads/s. Another purpose of our work supplemental to the verification of the Katz track theory for the dye dose meter, is to determine its response to very high doses as well as to radiation having an LET value significantly greater than those so far studied with this system.

3. BRIEF SUMMARY OF THE KATZ PARTICLE TRACK THEORY

The theory of Katz describes the response characteristics of detectors through the use of conventional target theory making use of models which have been called multi-hit and multi-target detectors. Further the theory involves the macroscopic dose response curve for low LET radiation for the determination of the response from a high LET ion by integrating the response of the energy profile of the ion over the range, which is affected by the delta rays generated by the ion. At this stage we have limited ourselves to single-hit processes, which is not a serious limitation, because in practice most detectors show response curves which may be interpreted as single-hit curves of radiation effect versus dose. In these detectors it is assumed that there are sensitive elements which require only a single radiation event or "hit" in order to produce the observed endpoint, which is the radiation damage we are looking for in our detector. Physically a "hit" may be interpreted as an event caused by an electron passing through the sensitive site with an effectiveness that depends on the speed of the electron. A one-hit detector is one in which it is assumed that there is a single target in which one hit can lead to the endpoint. From Poisson statistics it can be shown that such a detector always displays an exponentially saturating response as a function of dose, the relationship having the form $1-\exp(-D/D_{37})$, where D is the variable absorbed dose and D_{37} is the dose for 37% survival, the latter being characteristic of the type of radiation and the detector material. For such a detector the radiation response curve in a log-log plot has the same shape for different types of radiations and values of LET, but the numerical value of the D_{37} dose may be different from one detector to another.

Track theory is based on the assumption that the response to low LET radiations of different qualities can be adequately described by the energy imparted, namely the dose, and is essentially independent of the initial energy spectrum of photon and electron radiations. Furthermore we believe that this is due to the interactions leading to the detected endpoint occurring largely at the low-energy end of the slowing-down spectrum. In other words it is the δ-rays issued from the track spur which are mainly responsible for the deposited dose.

Gamma-ray photons and fast electrons deposit their energy through the secondary production of δ-rays, and therefore is the detector response to the same absorbed dose generally the same. But if the observed effect is not the same for different radiation qualities, the difference may be attributed to the rate at which the δ-rays are formed or to the spatial distribution of the δ-rays near the core of the particle track.

Heavy charged particles also deposit their energy by the formation of δ-rays, and on this basis the dose-response curve should show the same shape as for low-LET radiations. This is only true, however, as long as the effect from direct atomic and molecular interactions and displacements due to the heavy-ion interaction is negligible, and this means that the δ-ray theory of track structure cannot be used where the detected endpoint is mostly due to atomic and molecular interactions and displacements. As a test among many aspects of the track theory we would like to use a one-hit detector having a small sensitive site and a high D_{37} dose, which means a rather insensitive detector, and expose it to γ-rays and electrons of different energies, as well as heavy ions over a broad range of particle speeds and atomic numbers.

The dye film system satisfies the criterion of small sensitive-element size, since its response mainly is due to the interaction of electrons with single molecules and their immediate surroundings. The sensitive molecules themselves have a diameter of the order of 10 Ångstrøms. In addition the film is relative insensitive to ionizing radiation, which gives it a high D_{37} value.

4. PRELIMINARY EXPERIMENTAL RESULTS

The dose response curves for ^{60}Co γ-ray photons, 10 MeV electrons, and 16 MeV protons are shown in Fig. 2, and it is seen that the response at low doses is the same for the three different radiation qualities. At high doses a marked difference in saturation blackness takes place and a strong bleaching effect is observed. The difference in saturation blackness may come from competing reactions taking place, one being the forming of the highly coloured carbonium ion and the other a back reaction to the leuco form of the dye precursor. These deviations at high doses have not yet been studied, but they are unexpected and cannot completely be taken into account by the track theory.

For the heavy-ion irradiation one may consider the central path of a penetrating ion forming the axis around which radially ejected δ-rays transport the energy lost by the ion over transverse distances of many tens of microns, Fig. 3a. On their diverse paths each δ-ray deposits its energy by ionization and exitation of the molecules with a low dose. This dose response can be related to irradiation of the same material by low LET γ-rays, and we can write that the observed effect E is a constant times the dose:

$$E = k \cdot D$$

We have observed that the transfer coefficient k is the same for ^{60}Co γ-rays, 10 MeV electrons, and 16 MeV protons, at least for doses well below saturation of the radiation effect. Proportionality exists between this effect and the absorbed dose. The transfer characteristic is the same; the RBE for the dose meter is one. RBE for the dose meter refers to the reciprocal of the absorbed dose of a higher LET radiation to the absorbed dose of a low LET radiation, here γ-rays, required to produce the same observed effect in the dose meter.

But one may ask what happens in the center of the track core or very close to the core, where the ionization density must be very high? For heavy charged particles at least, saturation must exist and there will not be proportionality between the response and dose. Now looking at the macroscopic dose response for a high LET particle, the average observed effect will be less for the same amount of absorbed dose, and we can thus write the observed effect:

$$E < k \cdot D$$

A part of the deposited energy is wasted in this case, and the radiation effectiveness will therefore be smaller, and the RBE less than unity.

A qualitative estimation of the response characteristic for a high-LET particle for which the RBE in the film is less than unity will predict a dose response curve in a log-log plot being parallel to the response curve for γ-rays, having the same level of blackness at saturation, but having a higher D_{37} dose value, Fig. 3b.

5. CONCLUSION

For the actual experimental results as shown in Fig. 2, it is found that the saturation blackness is strongly dependent of the radiation quality, which means that the detector does not conform the ideal one-hit detector with respect to the saturation concept. On this background it is very difficult to make the full use of the Katz particle track theory as a quantitative determination of the response characteristic for high-LET radiation is impossible. More detailed investigations concerning the bleaching effect is necessary.

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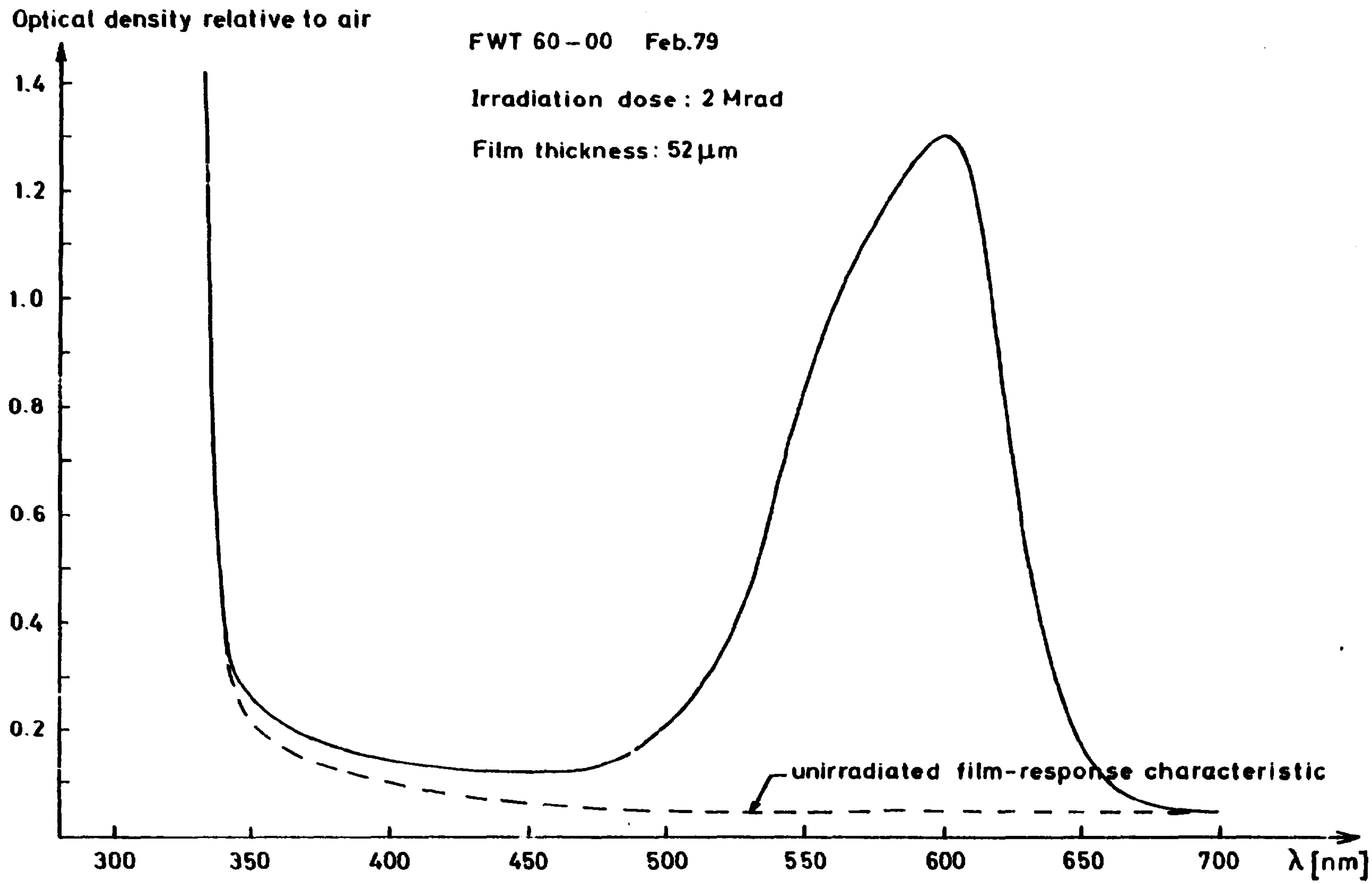
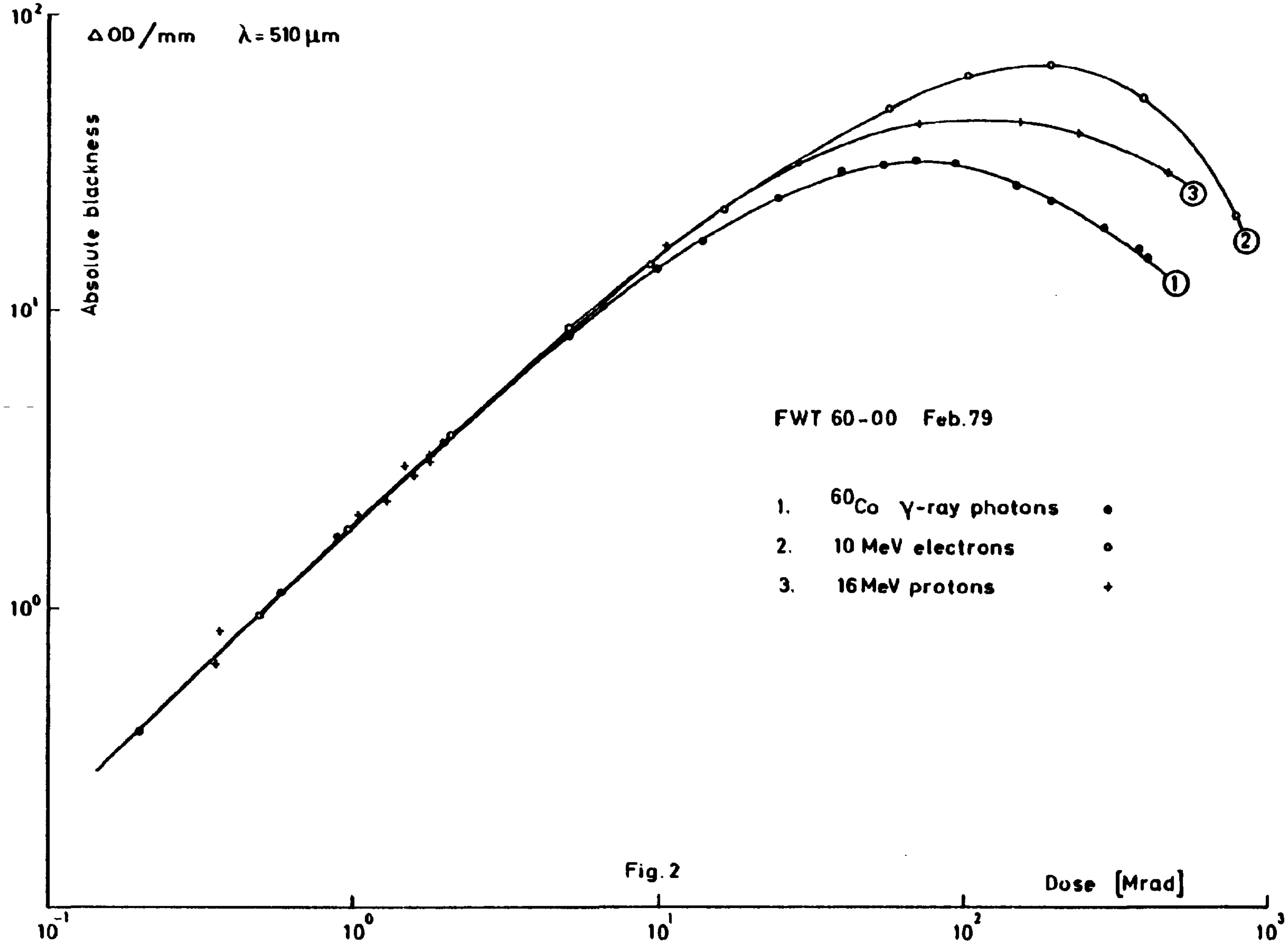

Optical density relative to air
FWT 60 – 00 Feb.79
Irradiation dose : 2 Mrad
Film thickness : 52 µm
1.4
1.2
1.0
0.8
0.6
0.4
0.2
unirradiated film-response characteristic
300
350
400
450
500
550
600
650
700
λ [nm]

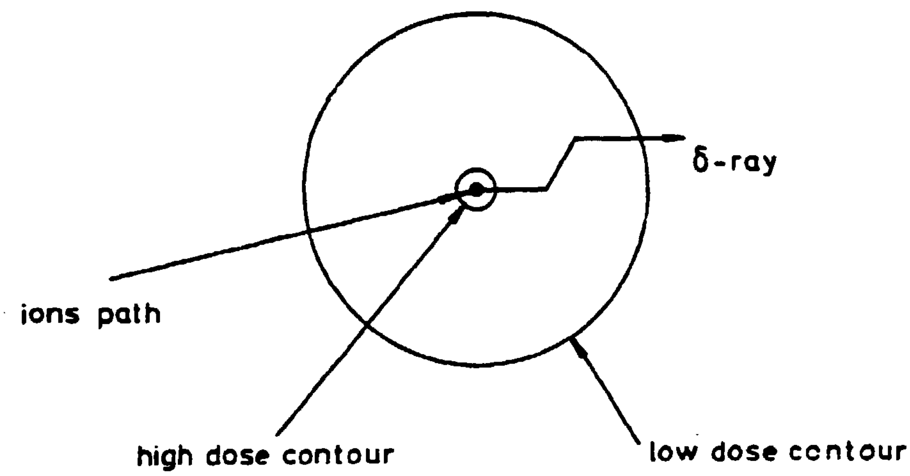
Fig 1.



10^2
Δ OD / mm $\lambda = 510\,\mu m$
Absolute blackness
10^1
10^0
FWT 60-00 Feb.79
1. ^{60}Co γ-ray photons •
2. 10 MeV electrons ∘
3. 16 MeV protons +
①
②
③
Dose [Mrad]
10^{-1}
10^0
10^1
10^2
10^3

Fig. 2

a. Microfield of the ion:



Low dose region:

measured effect $E = k \times dose$

k is a constant for low-LET radiations

b. High dose region:

measured effect $E < k \times dose$

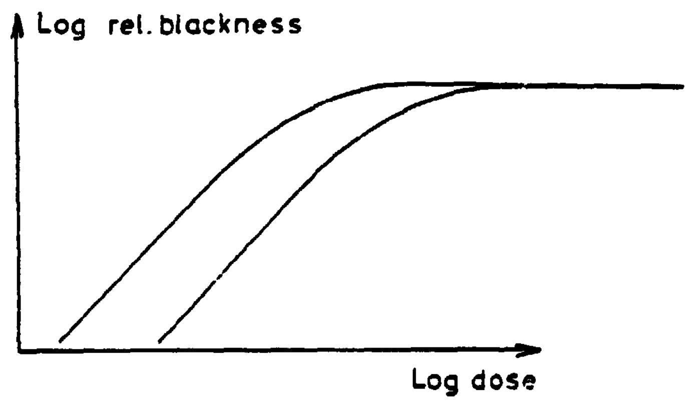


Fig.3

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Johnny W. Hansen

Date May 1980

Department or group

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