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THE RADIOCHROMIC DYE FILM DOSE METER AS A POSSIBLE TEST OF PARTICLE TRACK THEORY

Johnny W. Hansen, Mikael Jensen and Robert Katz

Abstract. The response characteristic of the thin-film radiochromic dye cyanide plastic dose meter to ionizing radiation of electrons and heavy charged particles is investigated as a possible test of the particle track theory worked out by Robert Katz and coworkers.

Dose response curves for low-LET radiation have been investigated and are used for a qualitative estimation of the response for protons and oxygen ions at 16 and 4 MeV/amu, respectively. A bleaching effect on the colouration at high doses intimates that the target cannot be interpreted literally, but it might still be possible to transfer the function of the macroscopic dose response to a theoretical dose response curve in a microscopic scale for a single ion. From this relation the macroscopic dose response curve can be determined when the film is irradiated with heavy ions.

It will be shown theoretically that for protons there is no saturation in the track core, whereas calculations for oxygen ions show a heavy saturation in the track core, which means that a part of the ions loose their energy ineffectively. We can conclude that it is possible qualitatively to pred. t the dose response curve for high-LET particles by means of the dose response curve for low-LET radiation.

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

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1. INTRODUCTION

The radiochromic dye cyanide film dose meter has been investigated as a possible test of the particle track theory of Katz. The radiation detector is a thin-film plastic dose meter developed for measurements of high absorbed doses and dose distributions in intense radiation fields [1-6]. The detector film is commercially available from Far West Technology, 330 Kellogg, Goleta, 93017 California, U.S.A. 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 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. The tested endpoint is the colouration, which we measure in terms of increase in optical density relative to air per unit film thickness (ΔOD/mm) at the wavelength of 510 nm, where the saturation optical density is about 3 for a film thickness of 5 mg/cm^2.

The track theory of Katz [7,8] describes the response characteristic of detectors from two independent hypotheses, namely conventional target theory making use of a model describing one-hit processes and the macroscopic dose response curve for low-LET radiation, e.g. γ-ray photons or fast electrons. The low-LET dose response curve is used to determine the response from a high-LET ion by integration of the response of the energy profile of the ion over the range which is influenced by the δ-rays generated by the ion. Gamma-ray photons and fast electrons deposit their energy through the production of δ-rays, and the detector response to equal absorbed doses is therefore in general the same. 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 radiation.

As a test of the track theory it is interesting to use a one-hit detector which has a small sensitive element site which is relatively insensitive to ionizing radiation. We have investigated the dye film dose meter to determine the extent to which it satisfies these criteria.

2. RELATIVE EFFECTIVENESS OF HEAVY IONS

For 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. On their diverse paths each δ-ray deposits its energy by ionization and excitation of the molecules. The dose response can be related to irradiation of the same material by γ-rays, and the observed effect is proportional to dose: Effect = K•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 RE for the detector is one. RE for the dose meter is defined as the ratio between the absorbed dose of low-LET radiation, γ-rays or electrons, and that of high-LET radiation required to produce the same observed effect in the detector.

For a heavy charged particle, however, where the ionization density in the track core and close to the core is sufficiently high, saturation must exist and there will be no proportionality between the response and dose. Now looking at the detector irradiated with such heavy charged particles, the average observed effect will be less for the same amount of absorbed dose, and we can thus write the observed effect for the detector as a whole as: Effect < K•D. A part of the energy deposited is wasted, therefore the radiation effectiveness is less and the RE is smaller than unity. The track theory will predict such ions which yield saturation in the track core and so have a RE less than one.

Assuming a uniform detector material with density ρ and irradiated with low-LET radiation to an average dose D, the energy E deposited in the sensitive element considered as a cylinder can be calculated from

$$E = D \cdot M_{cyl} = 2\pi a_o^3 \rho D$$

where M_{cyl} is the mass of the cylinder with the length of the axis equal to the diameter $2a_o$. Knowing the saturation dose D_o for γ-ray photons or fast electrons for that detector, the stopping power for a heavy charged particle which is able to saturate the sensitive element can be approximated:

$$\frac{dE}{dx_{a_o}} = \frac{E_o}{2a_o} = \pi a_o^2 \rho D_o$$

where dE/dx_{a_o} is the energy per unit length transferred to the cylinder volume. The track core is considered to coincide with the axis of the cylinder.

3. ARGUMENTS AGAINST A COMPLETE EXPLOITATION OF THE TRACK THEORY FORMULATION

Theory describes a one-hit detector by a response curve exponential up to saturation by the formulation $n = n_0(1-\exp(-D/D_{37}))$, where n is the volume density of the coloured dye molecules, n_0 is the total volume density of the leuco dye molecules, D_{37} is the radiation dose at which 63% of the leuco dye molecules are formed into the coloured dye molecules, and D is the independent variable dose. Comparing such a response curve normalized with respect to n_0 with an experimental determined response curve for the film irradiated with ^{60}Co γ-ray photons, fig. 1, it is found that the slope for the γ-ray photons follows a less steep response. Further when irradiating the film to very high doses above the saturation dose the response bends over and a strong bleaching effect takes place. At the moment it cannot be ruled out whether the bleaching effect always is present and so contributes to a less steep response also at low doses. If so the bleaching effect must be regarded as a competing effect to the formation of the coloured molecules, and we would then be dealing with two simultaneous effects whereas theory only takes one effect into consideration.

With the knowledge of the molar extinction coefficient [9] and the concentration by weight [2] of the leuco dye cyanide in the film, a theoretically achievable saturation optical density can be calculated from $OD = \varepsilon \cdot C \cdot l$, where ε is the extinction coefficient, C the concentration, and l the thickness of the film. The extinction coefficient is measured at the absorption maximum (601 nm) of the spectrum whereas our measurements are made off the peak where the optical density is lowered by a factor of five. At the actual wavelength of measurement the theoretical saturation optical density is calculated to have a value of 26, whereas the measured optical density for a film irradiated into saturation by ^{60}Co γ-ray photons has a value of 2.4. This small calculation indicates that about 10% of the leuco dye molecules are activated at saturation which again supports the assumption of competing reaction mechanisms taking place in the film material.

Irradiating the film into saturation with 10 MeV electrons and 16 MeV protons, response characteristics are obtained coinciding at low doses with the one obtained for ^{60}Co γ-rays. When saturation sets in a difference in the response is found and a marked difference in the saturation optical density exists, fig. 2. The track theory implies an exact quantitative value of the saturation optical density for a given detector, which means that the difference in saturation values must be noted when comparing response characteristics for high-LET particles where saturation in the track core exists. At this time we can offer no explanation for the difference in maximum optical density for the film irradiated with different radiation qualities. A more detailed investigation of the bleaching effect is necessary. The difference in saturation behaviour may be due to other aspects of radiation than radiation quality alone, e.g. dose rate and presence of oxygen.

Even if several competing processes exist it is reasonable to assume that these effects will be found from the δ-rays surrounding the path of high-LET particles. We conclude that competing effects take place upon irradiation of the detector which probably lead to a diminished utilization of the latent occurring leuco dye molecules. Based on the above mentioned difficulties a precise interpretation of the target must be omitted. Furthermore, in view of different maximum response levels only a qualitative description will be given. Discussion will be concentrated on proton tracks for which the center dose is very much lower and oxygen tracks for which the center dose is very much higher than the dose for saturation. We will assume that the response from low-LET radiation can be used to predict the response of high-LET radiation as described in the introduction.

4. ESTIMATION OF THE DOSE IN THE TRACK CORE

A quantitative estimation of the dose response curve for a high-LET particle relative to the one for γ-rays and fast electrons can be based upon calculations of the dose delivered to a small element of size of the dye molecule. In order to conform with earlier calculations of Katz [7,8] the element is given the form of a cylinder with the length equal to the diameter and placed coaxial to the path of the ion. We assume it appropriate to consider the size of the sensitive element as being the size of the dye molecule, as the energy deposited there can circulate in the symmetric rings of the molecule

[9]. In the track theory of Katz the D_{37} dose is related to the detector's dose response characteristic for γ-ray photons. For the actual detector we will use the dose response characteristic for fast electrons and the saturation dose D_o = 200 Mrad, but it will appear that the choice is not critical.

We may estimate the dose deposited by a 16 MeV proton in two ways. A first calculation may be based on stopping power. A 16 MeV proton penetrating a cylinder along the cylinder axis and giving up all its energy within the cylinder volume deposits a mean dose of

$$D_{pr} = \frac{\bar{E}_{pr}}{M_{cyl} \cdot k} = \frac{\bar{S}_{pr}}{\pi a_o^2 \cdot k} = 16 \text{ Mrad}$$

where $\bar{S}_{pr}$ = 32 MeVcm²g⁻¹ is the average proton mass stopping power in the film for the dye molecule, $a_o = 10^{-7}$ cm is the radius of the dye molecule, and $k = 6.24 \cdot 10^{13}$ MeVg^{-1}Mrad^{-1} is a conversion constant. The length of the small cylinder equals the diameter. However, a substantial fraction of energy is carried away by δ-rays so that the dose to these molecules lying in the center of the track is much lower than 16 Mrad. A second calculation utilizes the energy profile in fig. 3 calculated by Katz [7]. This shows that the dose in the center is about 6 Mrad. In either case the dose delivered to the dye molecule is very far from saturation, which occurs at 200 Mrad. Thus we see that the proportionality constant K discussed earlier should be the same for 16 MeV protons and fast electrons or γ-rays. In all parts of the track of the 16 MeV proton the dose to the dye molecule lies in the linear portion of its dose response curve.

For tracks of 4 MeV/amu oxygen ions a first calculation is based on stopping power. For a molecule lying in the track core and assuming that all the energy is absorbed in the molecule, the dose is calculated in the same way as for protons:

$$D_{oxygen} = \frac{\bar{E}_{oxygen}}{M_{cyl} \cdot k} = \frac{\bar{S}_{oxygen}}{\pi a_o^2 \cdot k} = 3.3 \cdot 10^3 \text{ Mrad}$$

where $\bar{S}_{oxygen}$ = 6521 MeVcm²g⁻¹ is the average oxygen-ion mass stopping power in the film for the dye molecule. The second calculation again utilizes the energy profile in fig. 3 calculated by Katz [7]. This shows that the dose in the center part of the core is about $1.6 \cdot 10^3$ Mrad which is 8 times higher than the saturation dose. Comparing these calculated doses we can see that half the energy is absorbed outside the molecule, and we can therefore conclude that about half the energy is delivered in a dose range of strong saturation. Thus we see that the proportionality constant K discussed earlier should be less than the constant valid for fast electrons or γ-rays.

5. DISCUSSION

Figure 4 shows in a linear plot experimental response values for ^{60}Co γ-rays, electrons, protons, and oxygen ions at low doses. As can be seen the response characteristic for protons has the same slope as the response characteristic for γ-rays and electrons, and so the 16 MeV protons have an effectiveness equal to the one for low-LET radiation in agreement with theory. The response characteristic for the 4 MeV/amu oxygen ions follows a less steep slope indicating a decrease in relative effectiveness by as much as 45% compared to the low-LET radiation, which corresponds qualitatively with our conclusion in the previous section that about 50% of the ions' energy is lost by oversaturation.

6. CONCLUSION

The present work has encouraged us to pursue the further applicability of the theory of track formation and to compare theoretical predictions of dose response characteristics with experimental results for high-LET radiation qualities. We have not yet shown experimentally how different high-LET radiation qualities will influence the film sensitivity, but we expect the effectiveness of heavy ions to depart gradually from that of γ-rays and electrons for increasing LET. Though the experimental results showed that the track theory of Katz could not be used on the whole, the qualitative interpretation of the results looked very promising for the use of the dye film as a detector in the test of the track theory.

A more intensive investigation is necessary in order to explain what is determining the different optical density values at saturation doses for different radiation qualities, and the reason for the dose response curve not to follow a one-hit response.

Finally our investigations have shown that it is possible to make use of microdosimetric examinations in order to derive macroscopic aspects of the dye film dose meter.

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Fig. 4. Optical density versus dose in a linear plot at low doses for ^{60}Co γ-rays, 10 MeV electrons, 16 MeV protons, and 4 MeV/amu oxygen ions.

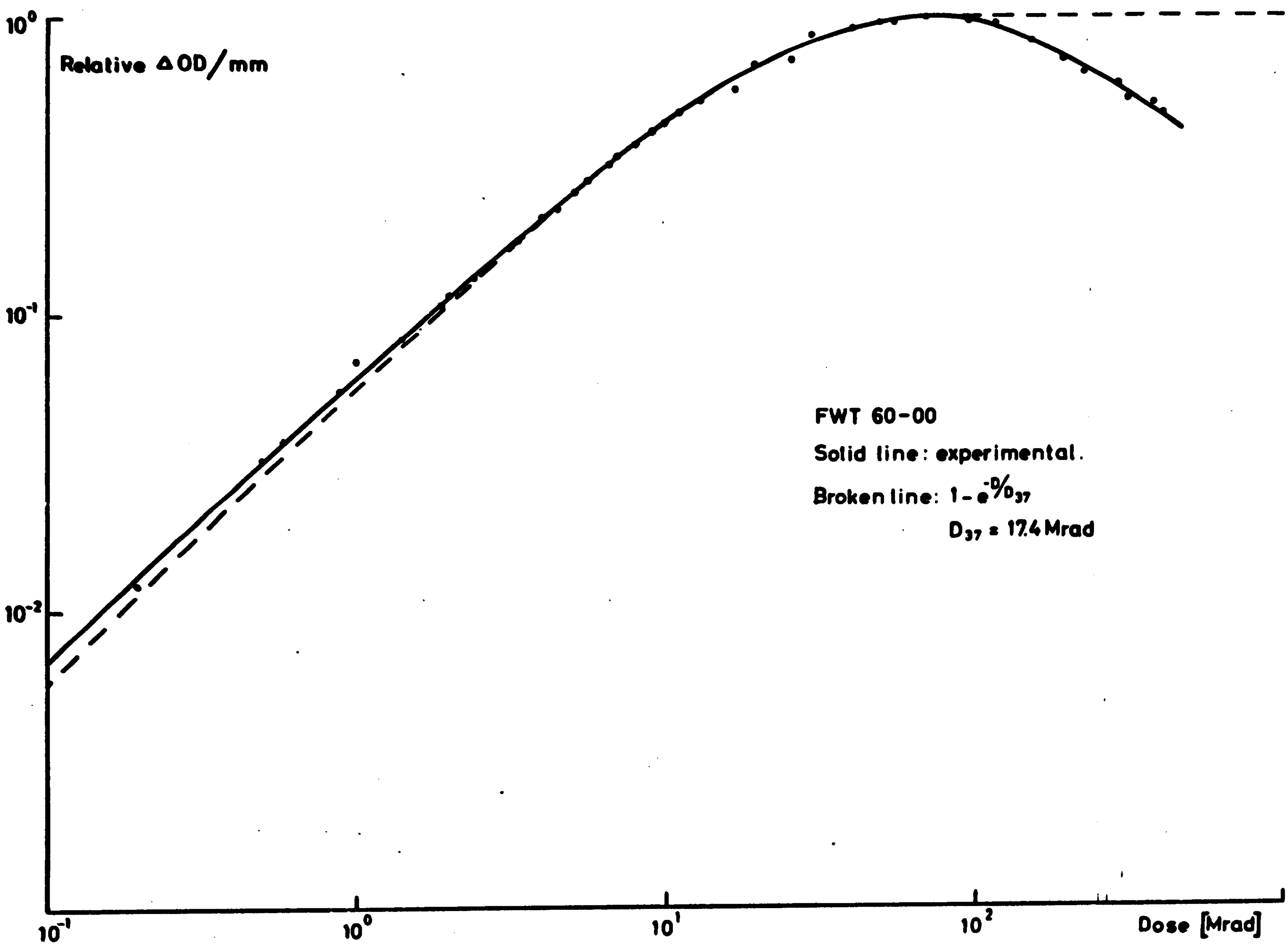
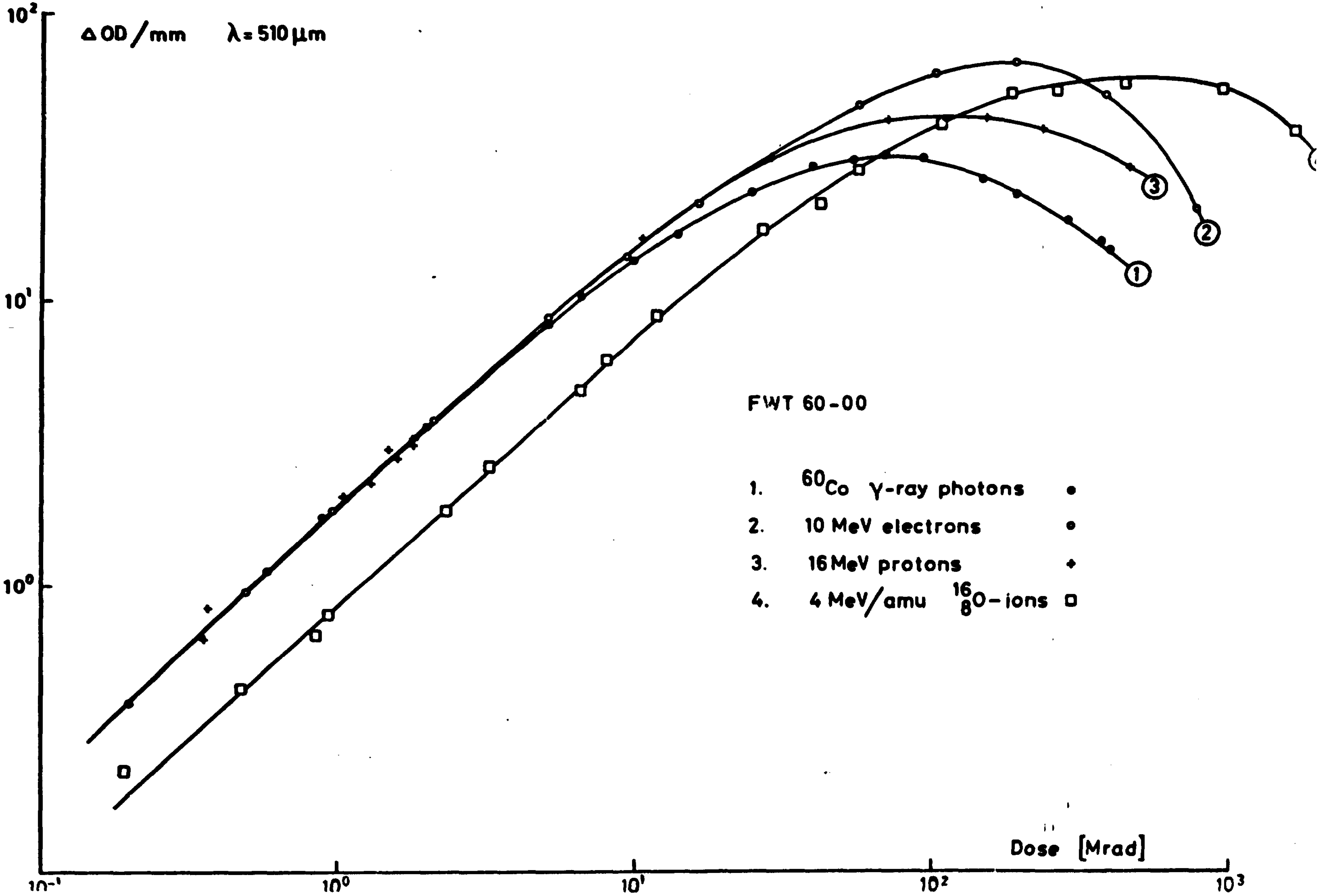

Relative ΔOD/mm
10^0
10^{-1}
10^{-2}
10^{-1}
10^0
10^1
10^2
Dose [Mrad]
FWT 60-00
Solid line: experimental.
Broken line: $1-e^{-D/D_{37}}$
D_{37} = 17.4 Mrad

Fig 1

Fig 2


ΔOD/mm λ=510 μm
10²
10¹
10⁰
10⁻¹
10⁰
10¹
10²
10³
Dose [Mrad]
FWT 60-00
1. ⁶⁰Co γ-ray photons ●
2. 10 MeV electrons ○
3. 16 MeV protons +
4. 4 MeV/amu ¹⁶₈O-ions □
①
②
③

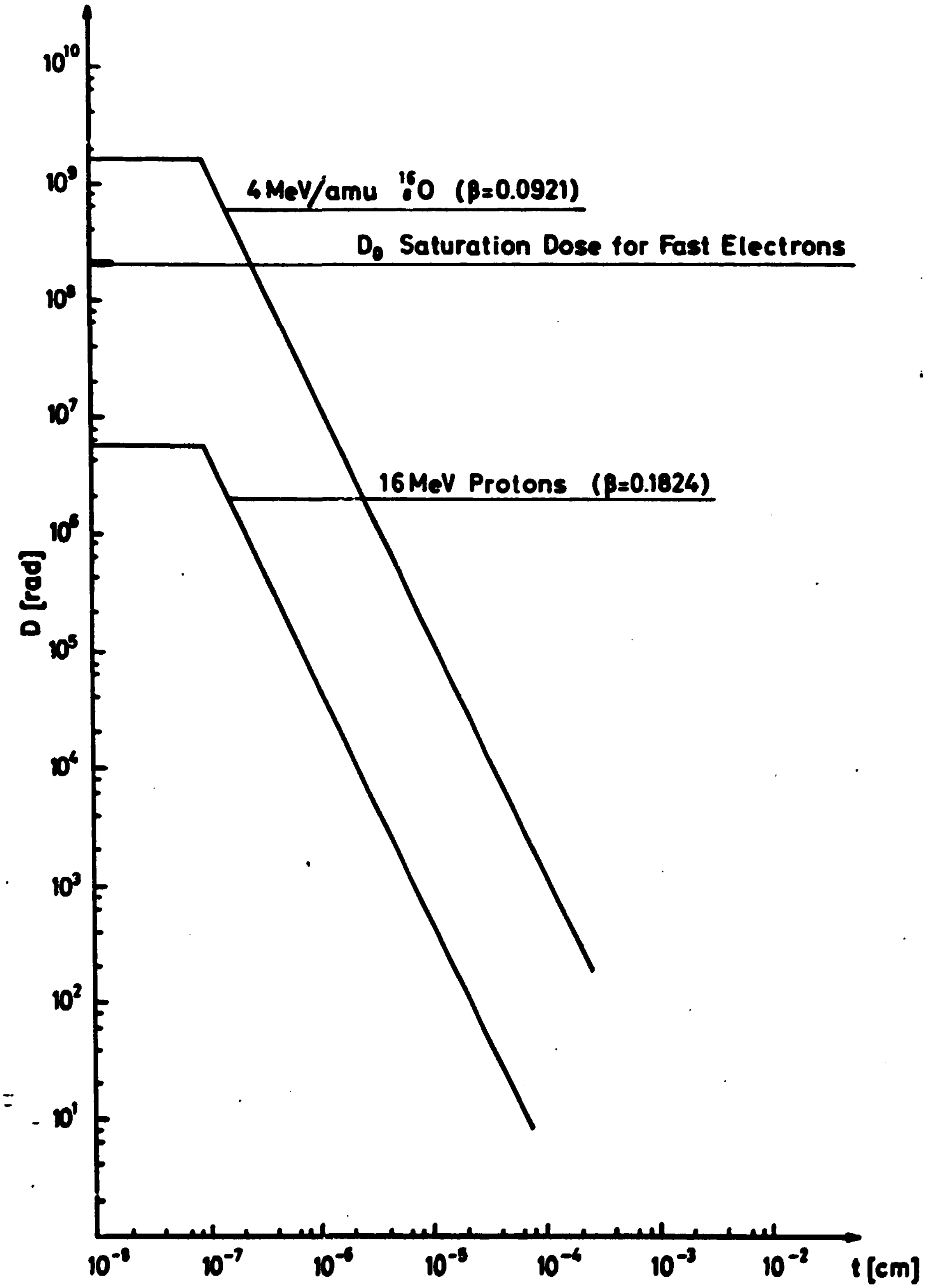
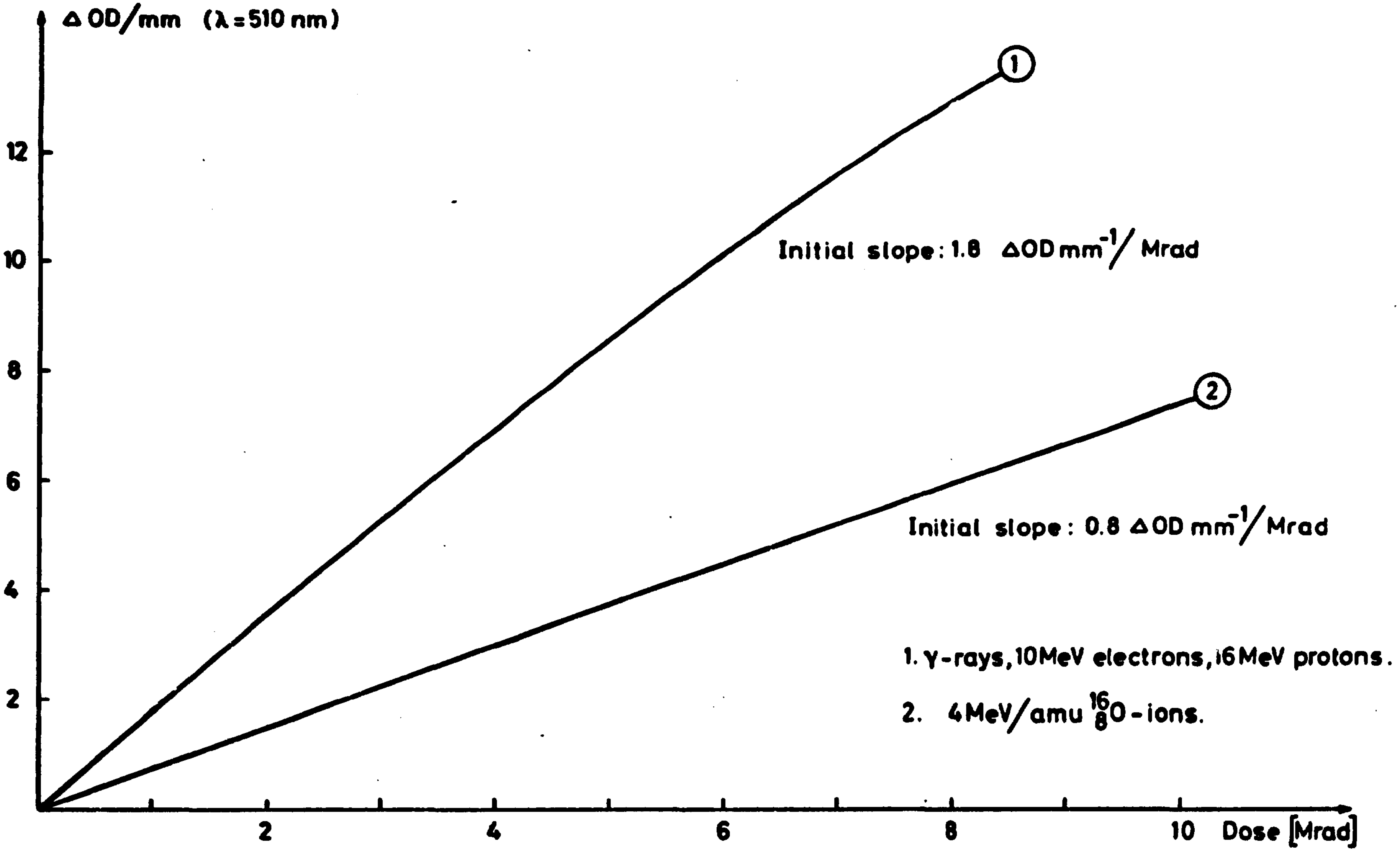

4 MeV/amu $^{16}_{8}O$ (β=0.0921)
D_0 Saturation Dose for Fast Electrons
16 MeV Protons (β=0.1824)
D [rad]
10^{10}
10^{9}
10^{8}
10^{7}
10^{6}
10^{5}
10^{4}
10^{3}
10^{2}
10^{1}
10^{-8}
10^{-7}
10^{-6}
10^{-5}
10^{-4}
10^{-3}
10^{-2}
t [cm]

Fig 3

Fig 4


ΔOD/mm (λ = 510 nm)
12
10
8
6
4
2
2
4
6
8
10
Dose [Mrad]
①
②
Initial slope: 1.8 ΔOD mm⁻¹/Mrad
Initial slope: 0.8 ΔOD mm⁻¹/Mrad
1. γ-rays, 10MeV electrons, 16MeV protons.
2. 4MeV/amu $^{16}_{8}$O-ions.

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