KERNFORSCHUNGSZENTRUM

KARLSRUHE

Juni 1976 KFK 2276

Abteilung Strahlenschutz und Sicherheit

New Approach in Megarad Dosimetry by Use of Coloured Cellulose Acetate

H. O. Mohamed

GESELLSCHAFT
FÜR
KERNFORSCHUNG M.B.H.

KARLSRUHE

GESELLSCHAFT FÜR KERNFORSCHUNG M.B.H.
KARLSRUHE

KERNFORSCHUNGSZENTRUM KARLSRUHE

KFK 2276

Abteilung Strahlenschutz und Sicherheit

NEW APPROACH IN MEGARAD DOSIMETRY BY USE OF COLOURED CELLULOSE ACETATE

H.O. Mohamed *)

*) on leave from the Egyptian Atomic Energy Authority

Gesellschaft für Kernforschung m.b.H. Karlsruhe

NEW APPROACH IN MEGARAD DOSIMETRY BY USE OF COLOURED CELLULOSE ACETATE

Abstract

Induced optical changes in transparent coloured cellulose acetate (green, blue, red,and yellow) have been investigated with respect to an application in radiation dosimetry.

It was found that the change in transmission gives better dosimetric properties than the change in optical density, and radiation response depends mainly on the colour of the sample and type of radiation. The useful dose range which can be covered by these coloured foils extends from 1 to 50 Mrad for e^-(10 MeV) and from 1 to 60 Mrad for ^{60}Co γ-rays.

Fading under laboratory conditions, the effect of temperature during storage time, the effect of UV radiation, the dose rate and energy dependence are also investigated. The results demonstrate the high reproducibility of dose measurement, with the coefficient of variation for electrons and γ-rays being of the order of 0.50 to 1.0 % for the dose range form 4 up to 50 Mrad. No significant variation was observed between different batches for the relative change of optical density.

NEUE WEGE DER DOSISMESSUNG IM MEGARAD-BEREICH DURCH VERWENDUNG VON GEFÄRBTEM ZELLULOSEAZETAT

Kurzfassung

In transparenten gefärbten Zelluloseazetatfolien (grün, blau, rot und gelb) induzierte optische Veränderungen wurden im Hinblick auf eine Anwendung in der Strahlendosimetrie untersucht.

Es zeigte sich, daß eine Veränderung der Transmission bessere dosimetrische Eigenschaften bietet als die Veränderung der optischen Absorption und die Strahlenempfindlichkeit hauptsächlich von der Farbe der Probe und von der Strahlenart abhängt. Der nutzbare Dosisbereich für die untersuchten Farbfolien reicht von 1 bis 50 Mrad für e^-(10 MeV) und von 1 bis 60 Mrad für ^{60}Co-Gammastrahlung.

Ebenfalls untersucht wurden das Fading unter Laborbedingungen, der Einfluß der Temperatur während der Lagerung, der Einfluß von UV-Strahlung, die Dosisleistungsabhängigkeit und die Energieabhängigkeit. Die Ergebnisse zeigen eine gute Reproduzierbarkeit der Dosismessung mit einem Variationskoeffizienten für Elektronen und Gammastrahlung von etwa 0,50 bis 1,0 % für den Dosisbereich 4 bis 50 Mrad. Für verschiedene Chargen wurden keine wesentlichen Unterschiede hinsichtlich der relativen Änderung der optischen Absorption beobachtet.

1. Introduction

A number of different solid materials having useful dosimetric properties have been reported in the literature making use of change of their colouration in the visible or ultraviolet wave length range during exposure to ionizing radiation. These materials fall into three main categories: glasses, single crystals and plastics. From the latter materials the dosimetric properties of coloured cellulose acetate foils will be described in this report. Increasing interest in plastics for dosimetric purposes in megarad range is closely related to the increasing number of large radiation sources used in radiation processing, radiosterilization, etc. Therefore during the past 25 years, many investigators have studied the properties of different kinds of plastic materials for dosimetry.
Day and Stein in 1951, first suggested the use of optical changes in plastics for dosimetric purposes [1]. They used a dyed type of polymethyl metharcrylate called "Perspex Red 400", the optical density increase at 600 nm being the measured parameter. The results indicated that oxygen-induced fading was a problem for the practical application. But, Whittaker [2] in 1964, and Orton [3] in 1966, improved this work and reported more informations about this material in detail.
Henley and Miller [4] in 1951, tried polyvinylchloride film plus a methyl-violet dye which was bleached at 600 nm, by the action of radiation due to liberated HCl. A useful range of 0.5 to 10.0 Mrad was found. The change in optical density was reported to be stable with storage time. Difficulties in sample preparation are the most draw back of this technique.

Henley and Richman [5] in 1954 and 1956 developed a very useful cellophane-dye system for dosimetry, which covers the range 0.5 to 15.0 Mrad. The bleaching of the 0.001 inch thick film at 655 nm is the measured quantity, and the increase in transmission is found to be a linear function of dose. The bleaching is permanent. Taimuty et al. [6] in 1958 reported that the film response is dose rate independent at least up to dose rates of 10^5 rad/s. Variability in the commerically available cellophane limits the reproducibility of dose indication to about 5 to 10 percent. Charlesby and Woods [7] in 1963 have investigated the response of this cellophane over a wider range of γ-doses, and compared the changes in colouration with those produced by α-particles. The results found with α-particles are more scattered and dependent on the penetration of α-particles through the cellophane.

Birnbaum et al. in 1955 first suggested using the UV absorption in plastics for dose measurements [8]. They performed a limited study on commercially available Melamine by measuring the optical density in the range 340 nm to 460 nm. From their results they predicted that the useable dose range can be extended from 5 x 10^2 to 1 x 10^7 rad by using different thickness from 0.1 to 1 cm. Results about dose rate dependence, fading, and precision were not reported.

Fowler and Day [9] in 1955, employed the UV absorption technique with polystyrene, demonstrating a useful dose range extending from one to at least 200 Mrad. The chief drawback reported was a large post-irradiation fading of the colouration, amounting to 50 % in 4 days. This limits the application to prompt exposure and readout situations.

Boag et al. [10] in 1958 also tried polystyrene in this method, but found it inferior to polymethylmethacrylate.

Artandi and Stonehill [11] in 1958 reported that rigid, non plasticized 0.01 inch thick polyvinylchloride film discoloured in the visible wavelength range after irradiation. The optical density at 400 nm was found to be a useful index of dose from 0.5 to 6 Mrad. Heat treatment of 4 hours at 120° C after irradiation was necessary to develop the colouration to a stable level, thus eliminating postirradiation instability. Both electron beams and γ-rays were measured successfully, but the dose rates used were not reported. Maul et al. [12] in 1961 have reported the performance of this material in greater detail.

Boag et al. [10] in 1958, throughly investigated polymethylmethacrylate as a dosimeter for high-intensity pulsed electron beams. UV absorption at 292 nm was found to increase linearity from 0.1 to 3 Mrad, independent of dose rate at least to 300 Mrad/s for pulses of 1 μs. The optical density usually increases by less than 5 % in 24 hours after exposure. This is followed by a slow fading of about 10 % per month. These fading is tolerable for most applications.

Davidson and Sutton [13] in 1964, Orton [14] in 1966, Berry and Marshall [15] in 1969, Chadwick [16-18] in 1969-1970 do more extensive study on clear perspex (PMMA) for gamma and electron measurements.

Boag et al. [10] in 1958, investigated UV absorption in Mylar as a dosimeter for high-intensity pulsed electron beams, particularly for the high dose range. A large 24-hour fading was observed, after which the remaining optical density

325 nm was stable for storge period of several months. Ritz [19] in 1961, found that this fading was due to oxidation and that no fading was observed in very thin Mylar (0.00025 inch). However, Ritz found that a dose rate dependence way present, so the system should be used with caution, particularly for pulsed or other nonconstant sources. The useful dose range extends from 5.0 to 1 000 Mrad.

Harris and Price [20] in 1961 found that optical density of polyvinyl vinylidene (Saran) at 260 nm vs. exposure was found to be a straight line in a log-log plot from 5×10^4 rad to 10^7 rad when irradiated with γ-rays. The change in absorption of Saran caused by irradiation is only slightly reduced when irradiated in vacume and measured in an inert atmosphere.

Rizzo [21] in 1969, performed some information for using clear transparent cellulose acetate butyrate as γ-ray dosimeter for high range.

Broszkiewicz and Bulhak [22] on 1973, reported that irradiation of clear transparent cellulose triacetate with γ-rays exhibits an absorption peak at 284 nm. The optical density vs. dose exhibits the ideal linear characteristic over the range 10 to 25 Mrad. The optical density is nearly, and even after prolonged storage, decreases by only a few percent. The reproducibility is of the order of ± 10 %.

Richold et al. [23] in 1973 investigated that thin film of polycarbonate (Makrofol 200 nm thick) can be used for γ-rays measurement for doses from 0.1 - 100.0 Mrad at wavelengths of 290 nm, 300 nm, and 325 nm. It has a precision of ± 2 % for doses from 1 to 50 Mrad. Only small changes occures during storage under a wide range of conditions. Above 37^oC the dosimeter is sensitive to temperature during irradiation and at dose rates below 50 Krad/h there is a reduction in response by about 20 %.

From the above review, up to now, one can find a lot of information about limited number of plastic dosimeters. (PVC., PMMA, and red perspex) which are used and establiched in many laboratories.
Although this types have some disadvantages, we found that since 20 years ago there was no interest to do more extensive studies and investigations of other types of plastics. The review of references have shown that coloured cellulose acetate was not used before for dosimetric purposes.
In this report, is coloured cellulose acetate introduced and investigated.

as a new high dose range dosemeter material for megarad dosimetry of gamma and electron radiations.

2. Experimental Techniques

2.1 Preparation of Samples

All transparent coloured cellulose acetate samples used are commercial materials not specialy intended for dosimetric purposes, which are produced and available from LONZA *) under trade name ULTRAPHAN.

Sheets of green, blue, yellow, and red transparent cellulose acetate with 0.200 mm thickness are cut into strips with standard size 2.5 x 1.5 cm, in order to fit conveniently into the sample holder of the spectrophotometer.

No attempts were made to dry or otherwise treat the samples by any means before or after irradiation, but it is preferable to wash the samples in distilled water, and drained on filter paper before each optical density measurement.

2.2 Methods of Irradiation

Gamma irradiations were performed in the Institute of Radiobiology of the Karlsruhe Nuclear Research Center with a ^{60}Co unit type 220 (0.5 Mr/h) which had been previously calibrated with Fricke ferreous sulphate dosimeter and other techniques.

Electron beam from the 10 MeV linear accelerator at the Federal Research Centre for nutration, Karlsruhe was used for electron irradiations. The average beam current was 320 μA, the pulse length 4.5 μsec, and the number of pulses per sec 175. The scan width was 40 cm. Based on the results of measurements with Fricke dosimeter the doses were calculated taking into account the energy used, beam current, the scan width, and conveyer velocity.
The samples were always irradiated in presence of air.

2.3 Optical Density Measurements

For all optical measurements, a Carl Zeiss spectrophotometer type PMQ II was used

---

*) Lonza-Werke GmbH, Weil am Rhein, Fed. Rep. Germany

with a holder devided into 4 devisions, so 3 samples plus the control one can be measured in one step.Clips are used to fix the samples flat during the measurement, so readings can be obtained with high reproducibility. The band width used was always less than ± 3 nm.

Scanning of the absorption spectrum of irradiated and unirradiated samples showed that the most suitable and convenient peaks for dosimetry purposes were 640 nm for green and blue samples, 560 nm and 490 nm for red and yellow samples, respectivly. All subsequent readings were made at this wavelenghts with respect to an unirradiated clear transparent sample of the same thickness for control. The initial optical density and transmission of the coloured samples are presented in Table 1.

sample colour	wavelength nm	Initial Optical Density	Initial Transmission %
green	640	1.36	4.5
blue	640	1.10	8.0
red	560	1.85	1.5
yellow	490	1.65	2.5

Tab. 1: Initial optical density and transmission for coloured cellulose acetate.

Optical density and transmission of each coloured sample with control was measured before and after irradiation at the appropriate wavelength.

3. Dose Characteristic

When coloured cellulose actetate is exposed to electrons or γ-radiation, bleaching of colour occurs. Consequently, optical density decreases and evantualy transmission increases. This decrease in optical density continue with increase of dose up to saturation. Normaly the amount of change depends on the kind of colour of the sample and the type of radiation. Typical absorption vs. dose curves are shown in Fig. 1 to 12. Figure 1 to Figure 4 presenting the change in optical density and in transmission respectively for the different coloured samples. These curves were obtained by irradiating a number of dosimeters (30 for each point) to a known dose level using the abovementioned ^{60}Co source and the 10 MeV linear accelerator.

The curves show that response of different coloured cellulose acetate to radiation is not the same, it differs with the kind of colour and the type of radiation. Instead of using the optical density, in many cases the change in transmission is better and preferable to be used as a quantity to determine the dose which is demonstrated in Fig. 5 to 12. The influence of colour is predominant for the dependance of the optical density on the dose. It may be seen that slopes of this curves differ considerable, which allow a sellection of the proper colour to measure the dose approximately linear over a large dose range.

4. Dosimetric Properties

4.1 Radiation Effects

High energy radiation induces several changes in plastic properies, most of which are a function of the total absorbed dose. Some plastics are dose rate dependent due to the influence of dose rate on free radical concentration. Also other changes in plastic are high enough to serve as a measure of radiation dose.

The common changes in plastics are those in appearance, in chemical or physical state, and in mechanical properties [24].

- Change in appearance occures showing temporary and permanent colour effects and bubbling.
- Chemical changes, include double-bond formation, dehydrochlorination, cross linking, oxidative degradiation, polymerization, depolymerization, and gas evolution.
- Physical changes, include effects on viscosity, solubility, conductivity, free radical spectra, fluorescence and crystallinity.
- Changes in mechanical properties include changes in tensile strength, elastic modules, hardness, elongation, flexibility, etc.

Irradiation of coloured cellulose acetate with ionizing radiation in air results in bleaching the dye for some extent. At high doses the rate of change

of optical density becomes smaller and appears to reach a limiting value at certain dose, according to the colour of dosimeter and type of radiation. This is to be expected due to the eventual complete destruction of the dye. Visual inspection shows that dosimeters irradiated to doses greater than this limiting value are in fact completly bleached due to either degradiation or to some other change in the bleaching dye.
Generaly, most of work done till now in colouration dosimetry make use of changes in optical density in the near UV or visible region as a function of dose. However, from our experience with coloured cellulose acetate, we notice that in many cases change in transmission gives better dosimetric properties than the change in optical density.

4.2 Batch to Batch Variation

The shape of the radiation-colouration wavelength band for coloured cellulose acetate depends primarly on the chemical structure of the dye stuff. It is to be expected that the general shape of the spectra, and the position of the peak, as well as the fading characteristic will not vary from one batch to another.While the shape of the irradiation-colouration band remains essentially constant from batch to another, the response that means ratio of the change in optical density and the absorbed dose, depends on dye concentration and therefore may be vary from batch to batch.

A comparison between samples from four different batches is given in Table 2 and 3, where each value of change in optical density is a mean of 10 readings after exposure to 10 Mrad.

It seems that there is no significant variation for the relative change of the optical density between the four batches.

sample colour	Batch No.	I.O.D	change in optical density	
			Δ.O.D.	$\frac{\Delta.O.D}{I\ O.\Delta}$
green	1	1.36	0.66	0.49
	2	1.30	0.62	0.48
	3	1.27	0.61	0.48
	4	1.18	0.55	0.47
blue	1	1.10	0.55	0.50
	2	1.00	0.51	0.51
	3	0.94	0.49	0.52
	4	0.90	0.48	0.53
red	1	1.90	0.66	0.35
	2	1.85	0.65	0.35
	3	1.80	0.64	0.36
	4	1.70	0.62	0.36
yellow	1	1.65	0.38	0.23
	2	1.60	0.36	0.23
	3	1.55	0.34	0.22
	4	1.50	0.33	0.22

Table 2: Influence of the initial optical density (I.O.D.) variation on the accuracy of dose measurement after 10 Mrad ^{60}Co gamma-irradiation for different batches of coloured cellulose acetate.

sample colour	Batch No.	I.O.D.	change in optical density	
			Δ.O.D.	$\frac{\Delta.O.D.}{I.O.D.}$
green	1	1.36	1.27	0.93
	2	1.30	1.23	0.95
	3	1.27	1.21	0.95
	4	1.18	1.14	0.96
blue	1	1.10	0.58	0.53
	2	1.00	0.53	0.53
	3	0.94	0.50	0.53
	4	0.90	0.49	0.54
red	1	1.90	1.12	0.59
	2	1.85	1.10	0.59
	3	1.80	1.07	0.59
	4	1.70	1.02	0.60
yellow	1	1.65	0.40	0.24
	2	1.60	0.37	0.23
	3	1.55	0.36	0.23
	4	1.50	0.35	0.23

Table 3: Influence of the initial optical density (I.O.D) variation on the accuracy of dose measurement after 10 Mrad irradiation with 10 MeV electrons.

4.3 Dose Range

The useful dose range which can be applied for all colours is shown in the Tables 4 to 7 for electrons and γ-radiation.

Samples became breakable after irradiation with γ-rays for more than 60 Mrad, for the investigations with electrons up to 50 Mrad no mechanical changes were observed.

Dose range Mrad	Changing parameter	Δ_2-Δ_1 *)			
		green	blue	red	yellow
1 - 30	optical	1.30	0.94	1.35	0.95
30 - 60	density	0.05	0.11	0.33	0.34
1 - 30	trans-mission	82	63	29	18
30 - 60	in %	11	23	37	23

*) change in the parameter corresponding to the dose range

Table 4: Change of optical density and transmission for different dose ranges (^{60}Co)

Dose range Mrad	Changing parameter	Δ_2 - Δ_1 *)			
		green	blue	red	yellow
1 - 10	optical	1.27	0.58	1.10	0.40
10 - 50	density	0.08	0.47	0.72	0.49
1 - 10	trans-mission	72	22	15	3
10 - 50	in %	22	56	76	12

*) change in the parameter corresponding to the dose range

Table 5: Change of optical density and transmission for different dose ranges (electrons, 10 MeV)

Dose range Mrad	Changing parameter	Priority in Response			
		1st	2nd	3rd	4th
1 - 30	optical density	red	green	yellow	blue
30 - 60		yellow	red	blue [1]	green [2]
1 - 30	trans-mission in %	green	blue	red	yellow
30 - 60		red	blue	yellow	green [3]

[1] saturation starts after 30 Mrad

[2] saturation starts after 40 Mrad

[3] saturation starts after 40 Mrad

Table 6: Coloured cellulose acetate samples arranged according to their priority for different dose ranges (^{60}Co).

Dose range Mrad	Changing parameter	Priority in Response			
		1st	2nd	3rd	4th
1 - 10	optical density	green	red	blue	yellow
10 - 50		red	yellow	blue	green [1]
1 - 10	trans-mission in %	green	blue	red	yellow
10 - 50		red	bluw	green	yellow

[1] saturation starts after 10 Mrad

Table 7: Coloured cellulose acetate samples arranged according to their priority for different dose ranges (electrons, 10 MeV).

4.4 Dose Rate Dependence

Generaly at very low dose rates it is to be expected that the effect of fading during the irradiation period would become significant for coloured cellulose acetate. No dose rate effect was detected within the range, 0.3×10^5 to 6×10^5 rad/h for ^{60}Co, and in the range 2×10^8 to 2×10^{10} rad/s for 5 MeV and 10 MeV electrons (see Figures 13 to 14). However, the different response found for ^{60}Co and electrons could be explained by a dose rate dependence.

4.5 Energy Dependence

Experiments conducted on plastics indicate that measured dose is nearly independent of the incident energy over a wide range. This is to be expected on theoritical grounds, since the energy absorbed is a function of the effective atomic number of dosimeter material [8,9,11]. Therefore the optical density measured in the plastic is approximatly proportional to the absorbed dose in tissue.
For electrons with energy 5 and 10 MeV, no difference was observed (Figure 15).

4.6 Surface Conditions

Due to the coloured samples dust particles collected on the samples shown only a small changes in optical density during measurment. This may be due to the fact that only a small number of the particles were in the measuring beam.
Finger prints on the film also showed a very little effect on the optical density.
When the surface of the sample was rubbed briskly with a kimwipe several times, resulting in small scratch marks, only change in optical density in the order of 5 % was observed.

5. Effects of Environmental Conditions

5.1 Fading

In this chapter we are discussing the fading of the dose indication under laboratory conditions during storage times up to periods of one month between

exposure and measurements. For irradiated coloured cellulose acetate, the behaviour of optical density with post-irradiation time is probably linked with the radiation induced chemical reaction products. The most important of this is bond breakage, oxidation, cross linking, polymerisation, free radical formation, double bond formation, etc. It is clear, however, that the distribution of bonds causes a continuing chemical reaction which takes place over long periods of time following irradiation.
The change in the optical density observed during storage periods in air is composed of two parts, a positive one where increase of optical density is due to colour centers formation, and a negative one where decrease of optical density is attributed to the effect of oxygen in air. This latter decrease is found by mathematical analysis to be influenced by diffusion, and is almost certainly due to oxygen penetrating the surfaces [14]. On this basis, the final equilibrium reached after certain time, is attributed to complete diffusion of oxygen into samples.
Post-irradiation behaviour of coloured cellulose acetate is mainly effected by type of radiation, the type of colours, dose range, and special environmental parameters.
Fig. 16 to 23 show typical fading curves for irradiated coloured cellulose acetate stored in laboratory conditions at temperatures of 20-25 ^{0} C, after irradiation to different doses of ^{60}Co gamma radiation and 10 MeV electrons. This figures demonstrate the high influence of the colour and the type of radiation on the shape of the fading curves. The smallest fading influence, even no fading for higher doses were found for yellow coloured cellulose acetate after gamma irradiation and for blue coloured cellulose acetate after electron irradiation. From all materials investigated, red coloured cellulose acetate shows the unfavourablest results due to a relative high decrease and increase, respectivly, of the optical density as a function of storage time and the amount of dose.

5.2 Temperature Effects during Storage

Change in optical density was investigated for samples which were irradiated with electrons and γ-rays for various doses and stored directly after irradiation in an oven at different temperatures for periodes up to 100 hours.

In the figure 24 to 55 the optical density of each coloured cellulose acetate foils is presented as a function of storage temperature up to 100^{0} C with the dose as parameter for each kind of detector, fading curves are given for storage periods of 1, 6, 24 and 100 hours at temperatures of 5, 25, 50, 70 and 100^{0} C.

Also the temperature fading demonstrate the high influence of the colour, of the radiation type and of the amount of dose on the decrease and increase, respectively, in the fading curves. For green coloured cellulose acetate (see Fig. 24 to 27 and 40 to 43) there is, for example, a decrease of the optical density at higher temperatures in the lower dose range, but an increase for higher doses especialy after electron irradiation. The smallest temperature fading was observed with blue coloured cellulose acetate (see Fig. 48 to 51) after electron irradiation, contray to the significant decrease of the optical density for higher temperatures after gamma irradiation (see Fig. 32 to 35). The yellow cellulose acetate on the other side shows the lowest change in the optical density after gamma irradiation (see Fig. 28 to 31), but an increase as a function of storage temperature especialy after high dose of electron irradiations (see Fig. 44 to 47). For red cellulose acetat the optical density after high dose gamma irradiation seems to be constant up to a temperature of 50^{0} C followed by a rapid decrease at higher temperatures (see Fig. 36 to 39). The same detector shows an increase of the optical density for high doses of electrons above a storage temperature of 50^{0} C (see Fig. 52 - 55).

Samples irradiated with gamma rays for more than 40 Mrad and stored directly after irradiation for more than 24 hours in an oven at 100^{0} C, became more brittle with some increase in hardness. Contrary to this results we found no change in the mechanical properties after exposure to electrons of 50 Mrad and a treatment under the above conditions. It should be pointed out that these changes depend on radiation dose, kind of radiation, storage temperature, storage time and colour of sample.

5.3 UV Effects

When samples of coloured cellulose acetate were irradiated with electrons and gamma radiation for different doses, and directly after irradiation exposed to UV for different periods, change in optical density was observed. Of course the magnitude of this change depends on the order of previously electron and gamma irradiation, time of UV exposure, and the colour of sample (Fig. 56 to 63).

Generaly, optical density decrease in most cases as a function of UV exposure,

while it is slightly increase in few cases after longer exposure to electrons (see Fig. 60 and 61) as well as in the first exposure period for gamma irradiation (see Fig. 57). The yellow coloured cellulose acetate show the smallest response to UV light.
No mechanical changes were observed for samples irradiated with electrons to UV up to 24 hours. But little increase in hardness was observed for irradiation with gamma rays under the same conditions.

5.4 Day Light Effects

For irradiated samples (with electrons or gamma rays), change in optical density is approximately the same, whether they are stored in dark or day light. But it is clear that exposure directly to strong sunlight for long time could result in temperature effect due to the influence of UV light (see chapter 5.3) and heat.

6. Reroducibility of Dose Measurement

The standard error inherent in the method itself,i.e error of repeated measurements on the samples of cellulose acetate under the same conditions, has been determined experimentaly at several dose levels by irradiating groups of fifty samples of each colour with electronsand gamma radiation, and subsequantly for each group receiving the same dose, the mean value as well as the standard deviation of changing in optical density were defined.

The analysis of this results are given in Tabels 8 and 9. It appears that the error is relatively higher at low doses (because bleaching of dye occures and not darking), where optical density measurement seems to be less accurate compared to the cange in transmission.
It was found that the coefficient of variation is in the order of o.50 to one % after irradiation with electrons to doses of more than 2, 6, and 10 Mrad corresponding to green, blue and red, and yellow colour, also and after irradiation with γ-rays to doses of more than 4, 8, 10 Mrad corresponding to green, blue, red and yellow coloured cellulose acetate, respectively,
The results demonstrate the high reproducibility of dose measurements in the megarad range even for doses up to 50 Mrad in comparison with the other results reported.

Dose Mrad	coefficient of variation in %			
	green	blue	red	yellow
1	3.47	6.28	3.08	8.00
2	1.95	2.38	2.58	6.30
4	1.00	1.96	1.50	2.93
6	0.92	1.39	1.10	2.04
8	0.85	1.06	0.97	2.06
10	0.74	0.72	0.88	1,75
20	0.54	0.45	0.34	1.03
30	0.45	0.44	0.42	0.71
40	0.44	0.43	0.35	0.62
50	0.45	0.43	0.31	0.54

Table 8: Reproducibility of coloured cellulose acetate at different doses (electrons, 10 MeV)

Dose Mrad	coefficient of variation in %			
	green	blue	red	yellow
1	4.00	3.50	7.50	9.00
2	3.00	2.50	6.00	7.50
4	2.10	2.00	4.50	3.50
6	1.00	1.50	3.20	2.45
8	0.90	1.20	2.00	2.15
10	0.75	0.90	1.67	1.90
20	0.45	0.50	1.10	0.65
30	0.35	0.40	0.85	0.50
40	0.32	0.35	0.75	0.40
50	0.30	0.35	0.60	0.35
60	0.30	0.33	0.55	0.30

Table 9: Reproducibility of coloured cellulose acetate at different doses (^{60}Co).

7. Conclusion

Transparent coloured cellulose acetate can be used simply as a megarad dosimeter for electrons and γ-rays for dose range 1 to 50 and 1 to 60 Mrad, respectively. This appears in table 6 and 7 where the priority in response of different colours with radiation are demonstrated.

It was found that radiation response depends mainly on the colour of sample and radiation type. For good dosimetric properties the use of a change in transmission is better than a change in optical density (Fig. 1 to 12).
Fading results at laboratory conditions show smallest fading, even no fading for higher doses by using yellow samples after γ-irradiation and by using blue samples after electron irradiation, while red samples show the unfavourablest results for both electrons and γ-rays (Fig. 16 to 23).

Temperature fading results demonstrate the high influence of the colour, the type of radiation and radiation energy, respectively, and the amount of dose on the decrease and increase of the induced optical density (Fig. 24 to 55).

Exposure of irradiated samples to UV decreases the optical density in many cases, while it is slightly increase in few (Fig. 56 to 63).

Reproducibility of dose measurement was analysed in table 8 and 9 showing that the error is relativly lower at high doses where the coefficient of variation was found to be approximately 0.4 %.

Acknowledgment

I should like to express my sincere thanks to Mr. E. Piesch for his valuble scientific discussions and guidance during these work. The help of Mr. B. Burgkhardt who assisted in solving many scientific and technical problems was most welcome. Thanks are also to Dr. Grunewald, Mrs. R. Richter and Mr. M. Rudolf of the Federal Research Centre for Nutration, Karlsruhe, for dose estimation and sample irradiations. I thank also Mr. E. Lachmann for the preparation of the figures and Mrs. H. Kompalla for her assistance in typing.

This work was supported partially by an IAEA fellowship and partially by a fellowship within the German-Egypt Cooperation Project between KFA-Jülich and the Egyptian Atomic Energy Commission.

References

[1] M.J. Day, G. Stien: Effects of x-rays upon plastics, Nature, 168, 644 (1951).

[2] B. Whittaker: Radiation dosimetry technique using commercial red perspex: Report, AERE-R 3360, 1964

[3] C.G. Orton: Red perspex dosimetry, Phys. Med. Biol., II, 551 (1966)

[4] E.J. Henley, A. Miller: Gamma ray dosimetry with polyvinylchloride films, Nucleonics, 9 (No. 6), 62 (1951)

[5] E.J. Henley, D. Richman: Cellophane-dye dosimeter for 10^5 to 10^7 roentgen range, Anal. Chem., 28, 1580 (1956)

[6] S.I. Taimuty, R.A. Glass, B.S. Deaver: High level dosimetry of gamma and electron beam sources, Proceedings of the second International Conference on peaceful uses of atomic energy, 21, 204, Geneva (1958)

[7] A. Charlesby, R.J. Woods: Radiation dosimetry with dyed cellophane, Int. J. Appl. Rad. Isotopes, 14, 413 (1963)

[8] M. Birnbaum, J. Schulmann, L. Seren: Use of Melamine as an x-radiation detector, Rev. Sci. Instr., 26, 457 (1955)

[9] J.F. Fowler, M.J. Day: High dose measurement by optical absorption, Nucleonics, 13 (No. 12), 52 (1955)

[10] W.J. Boag, G.W. Dolphin, J. Rotblat: Radiation dosimetry by transparent plastics, Rad. Res. 9, 589 (1958)

[11] C. Artandi, A.A. Stonehill: Polyvinylchloride - new high level dosimeter, Nucleonics, 16 (No. 5), 118 (1958)

[12] J.E. Maul, N.W. Holm, I.G. Draganič: The use of polyvinylchloride film for Co-60 radiation dosimetry, Riso Report No. 31, 1961

[13] G. Davidson, H.C. Sutton: Commerical 1 mm clear perspex as a gamma-radiation dosimeter, Int. J. Appl. Rad. Isotopes, 15, 74 (1964)

[14] C.G. Oyton: Clear perspex dosimetry, Phys. Med. Biol., II (No. 3), 377 (1966)

[15] R.J. Berry, C.H. Marshall: Clear perspex H.X. as a reference dosemeter for electron and gamma radiation, Phys. Med. Biol., 14 (No. 4), 585 (1969)

[16] K.H. Chadwick: The use of PMMA as a dosimeter for irradiation studies in the range 10 Krad to 10 Mrad, Atompraxis, 15 (No. 3), 181 (1969)

[17] K.H. Chadwick: An interpretation of the optical density - dose relationship in irradiated clear PMMA, Rad. Res., 44, 282 (1970)

[18] K.H. Chadwick: The choice of measurement wavelength for clear, HX-perspex dosimetry, Proc. IAEA-Symp. Dosimetry in Agriculture, Industry, Biology, and Medicine 1973.

[19] V.H. Ritz: A note on Mylar Film dosimetry, Rad. Res, 15, 460 (1961)

[20] K.K. Harris, W.E. Price: A thin plastic radiation dosimetry, Int. J. Appl. Rad. Isotpes, II, 114 (1961)

[21] F.X. Rizzo: Cellulose acetate butyrate dosimeter, Trans., 12, 61 (1969)

[22] R.K. Broszkiewicz, Z. Bulhak: Plastic film dosimeters, Proc. IAEA-Symp. Dosimetry in Agriculture, Industry, Biology, and Medicine 1973

[23] P.H.C.V. Richold, J.A. Douglas, M. Marshall, J.A.B. Gibson: A megarad plastic film dosemeter, Phys. Med. Biol., 18 (No. 5), 665 (1973)

[24] J.W. Ryan: Effect of gamma radiation on certain rubbers and plastics, Nucleonics, II, No. 8, 13 (1953)

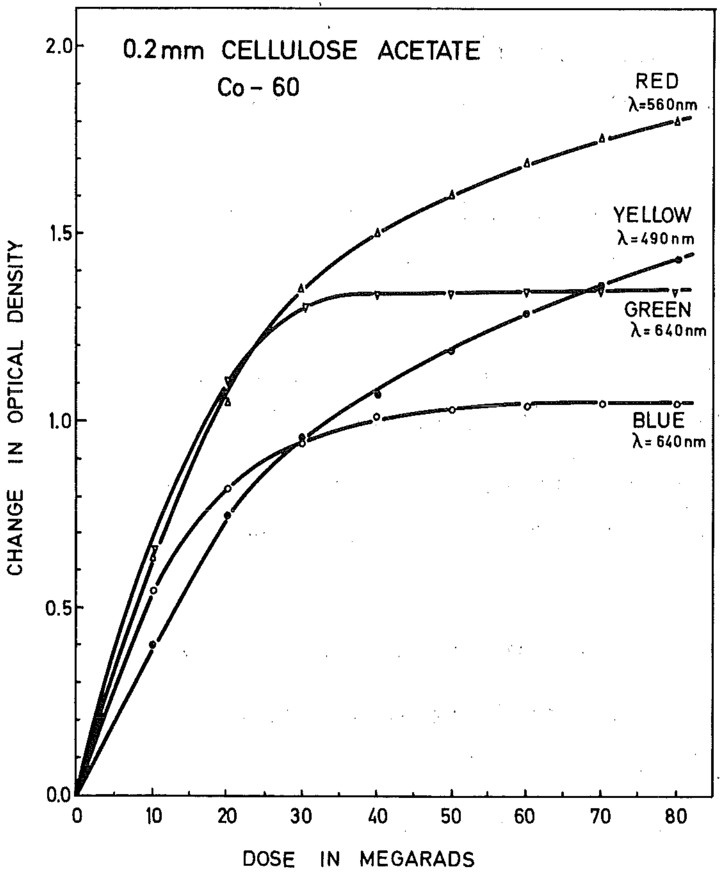

Fig. 1:

Change in optical density of coloured cellulose acetate irradiated with γ-rays

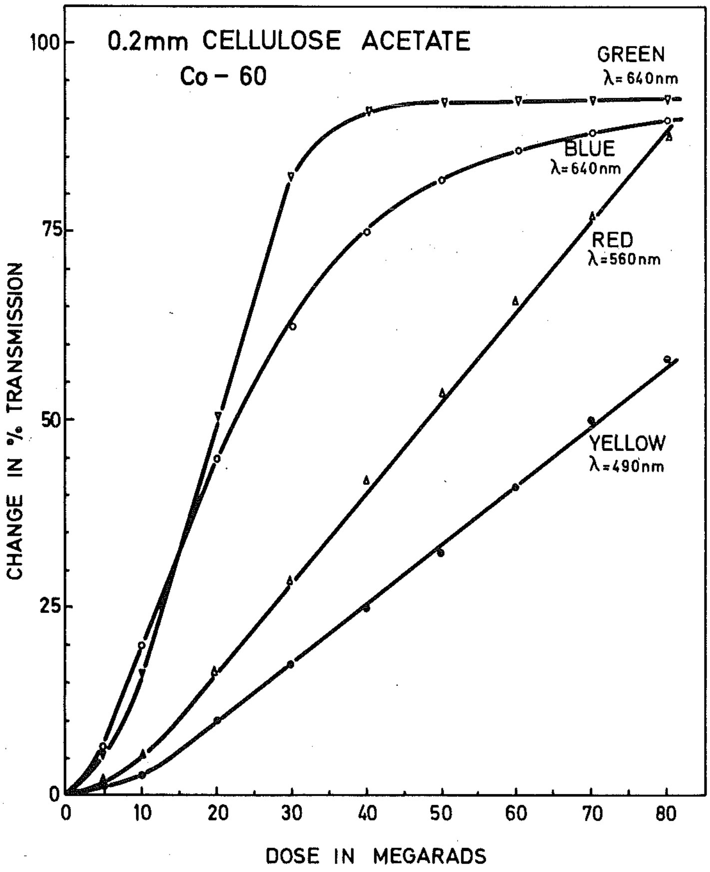

Fig. 2:

Change in % transmission of coloured cellulose acetate irradiated with γ-rays

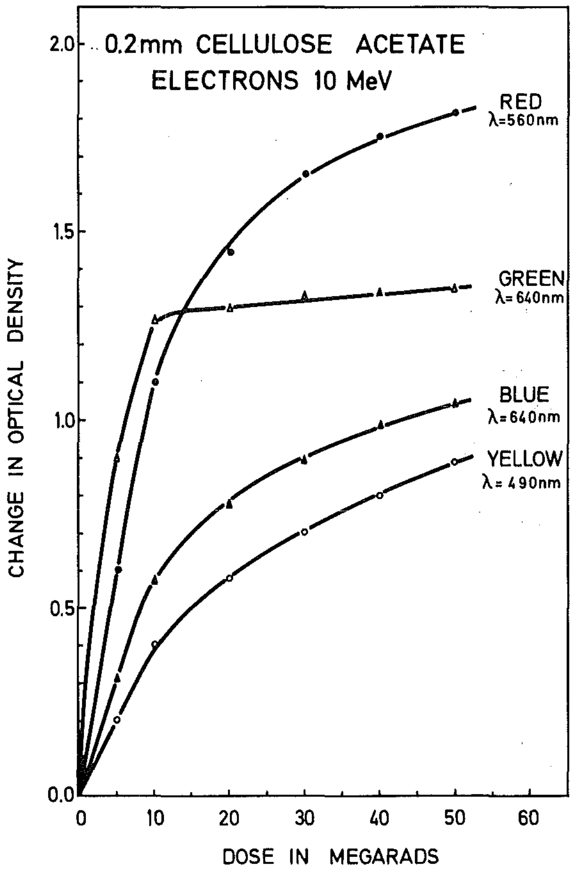

Fig. 3: Change in optical density of coloured cellulose acetate irradiated with electrons

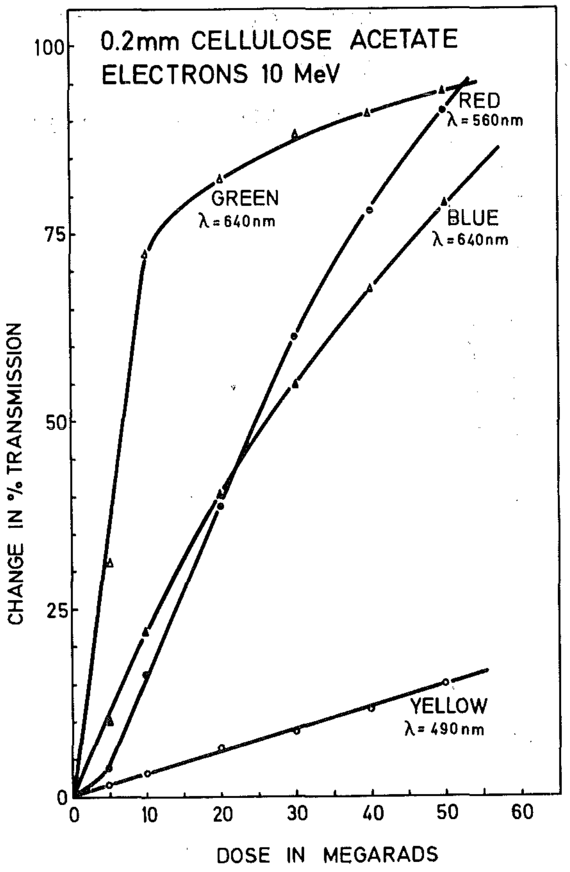

Fig. 4: Change in % transmission of coloured cellulose acetate irradiated with electrons

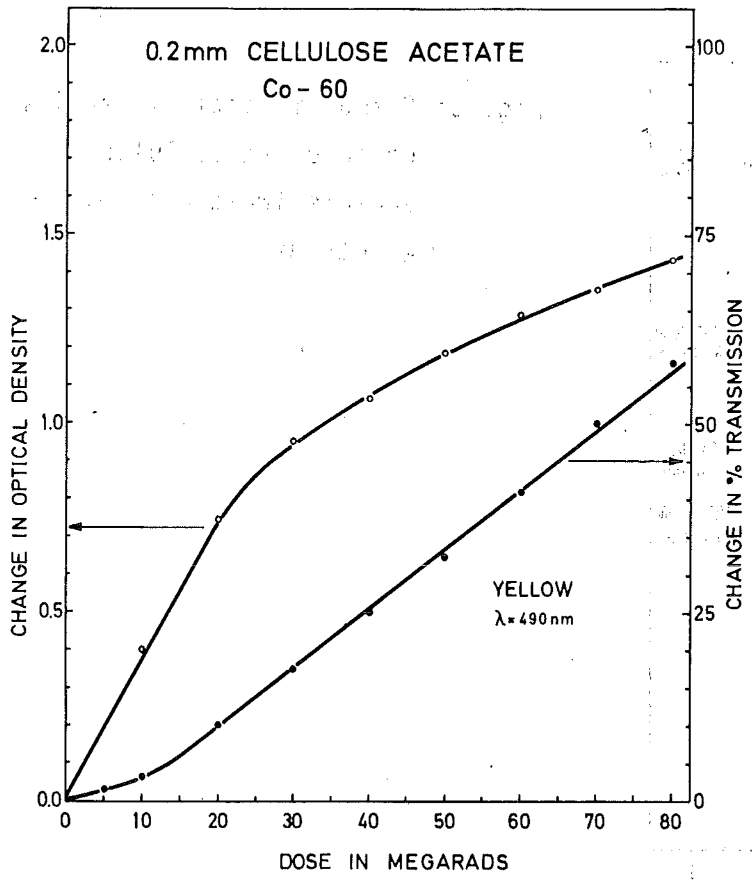

Fig. 5:
Dose response of yellow cellulose acetate irradiated with γ-rays

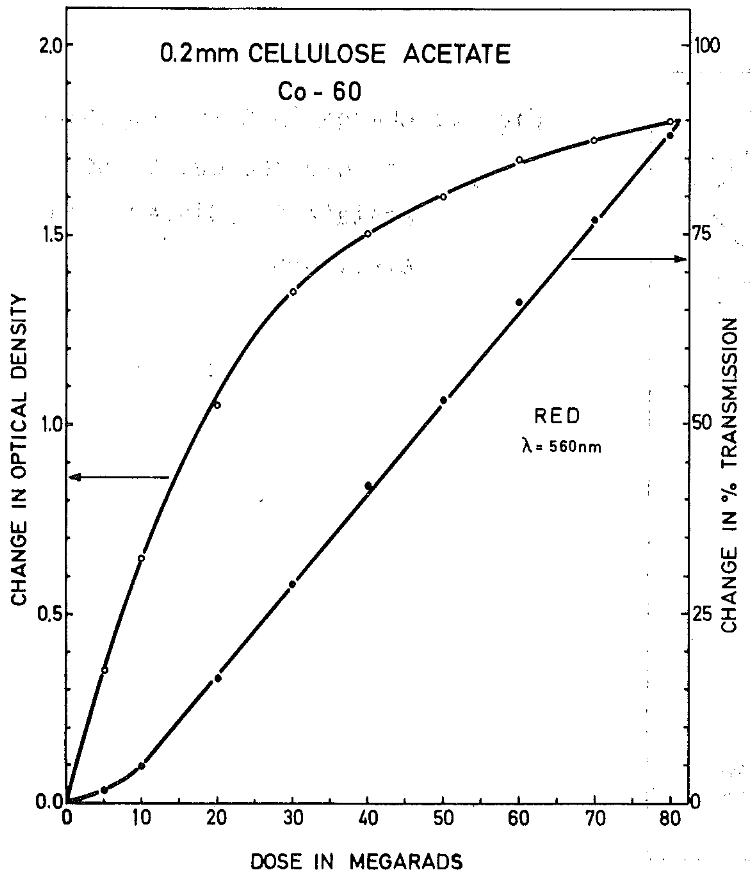

Fig. 6:
Dose response of red cellulose acetate irradiated with γ-rays

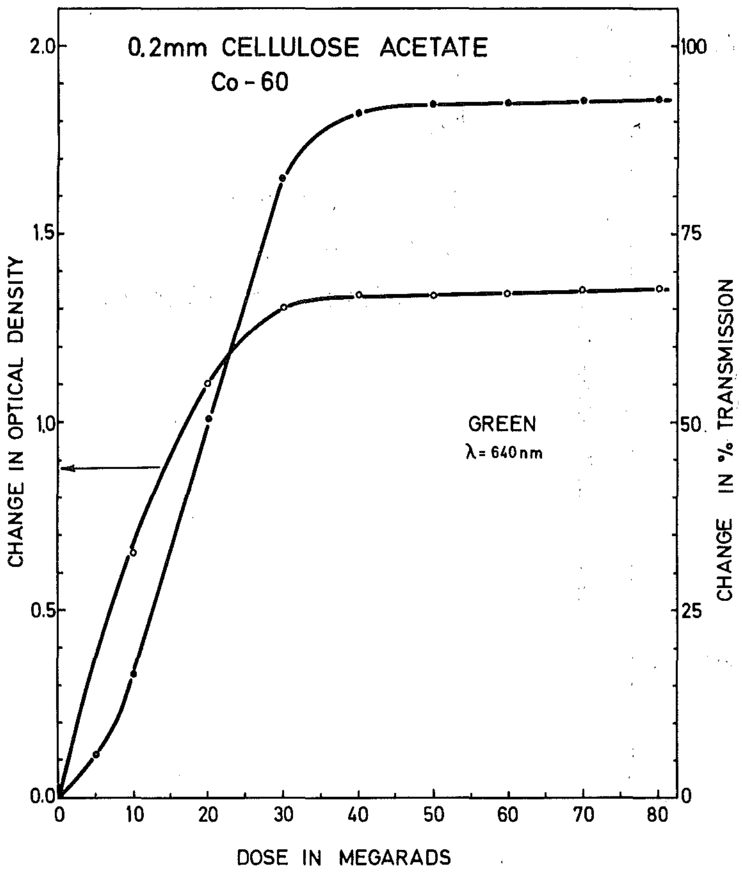

Fig. 7:

Dose response of green cellulose acetate irradiated with γ-rays

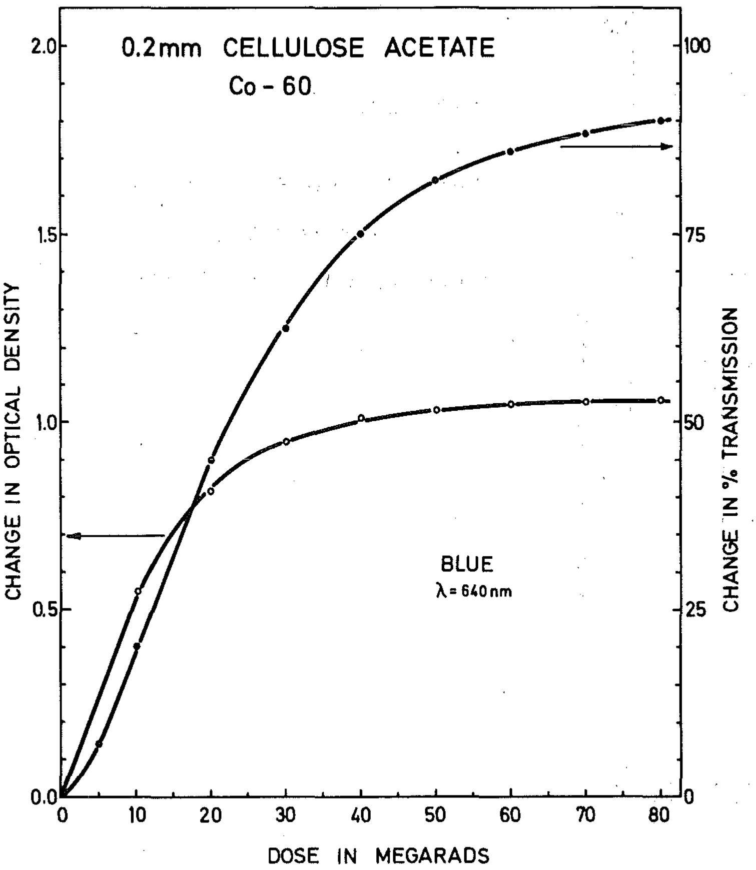

Fig. 8:

Dose response of blue cellulose acetate irradiated with γ-rays

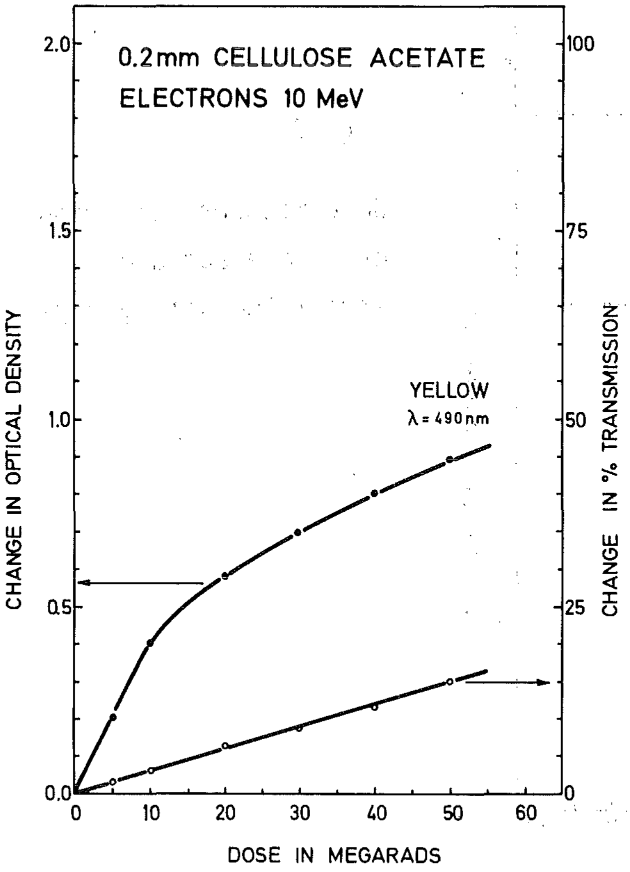

Fig. 9:

Dose response of yellow cellulose acetate irradiated with electrons

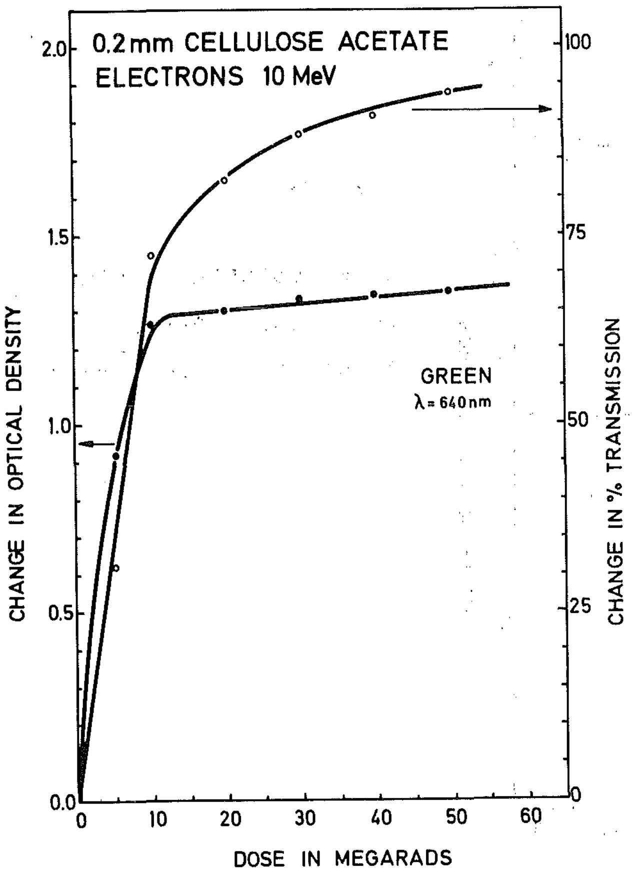

Fig. 10:

Dose response of green cellulose acetate irradiated with electrons

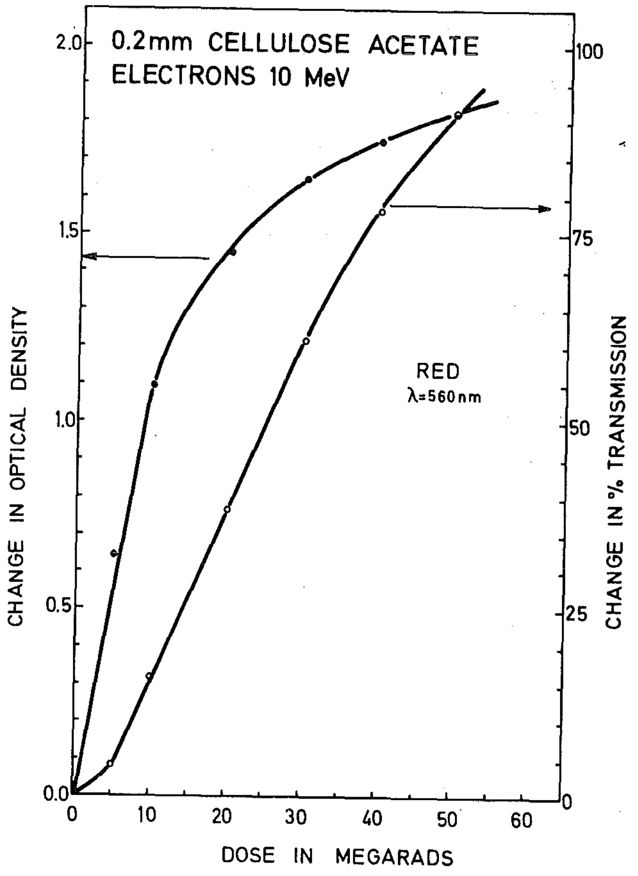

Fig. 11:

Dose response of red cellulose acetate irradiated with electrons

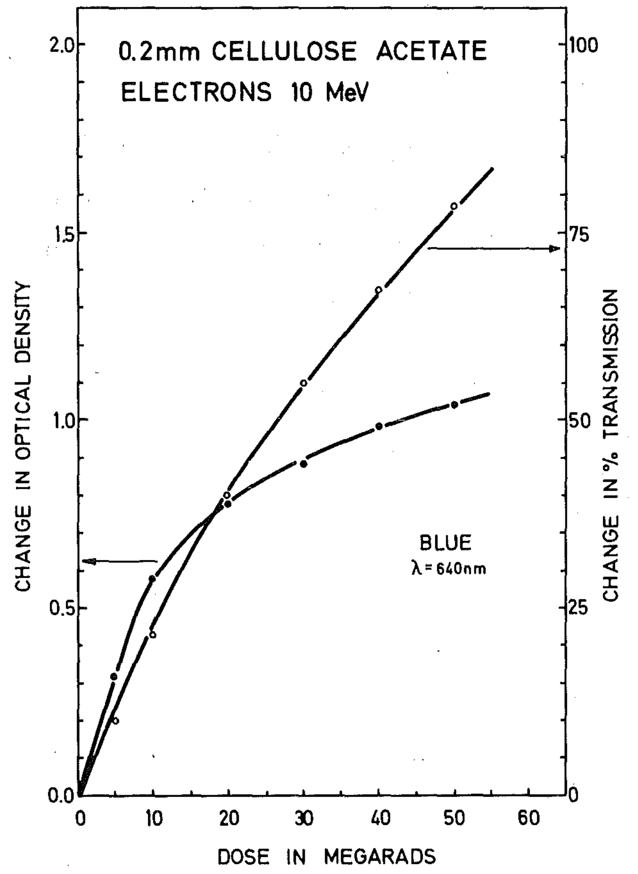

Fig. 12:

Dose response of blue cellulose acetate irradiated with electrons

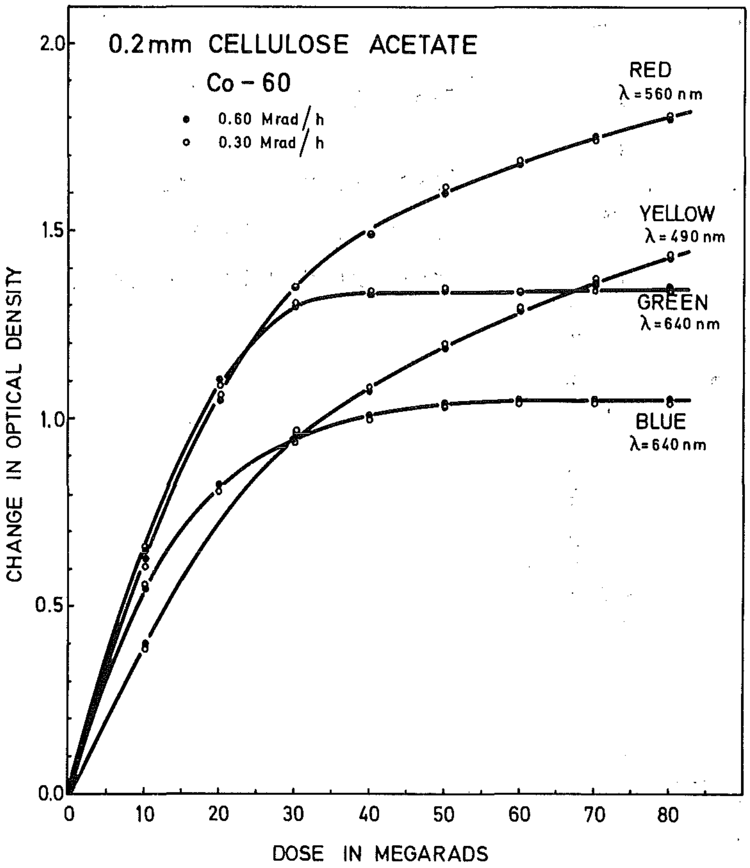

Fig. 13:

Dose response of coloured cellulose acetate irradiated with γ-rays

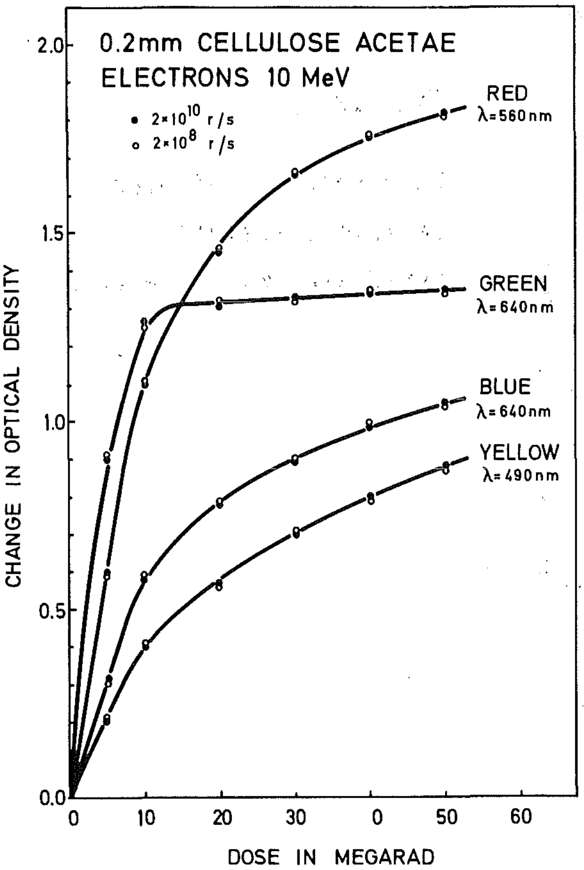

Fig. 14:

Dose response of coloured cellulose acetate irradiated with electrons of different dose rate

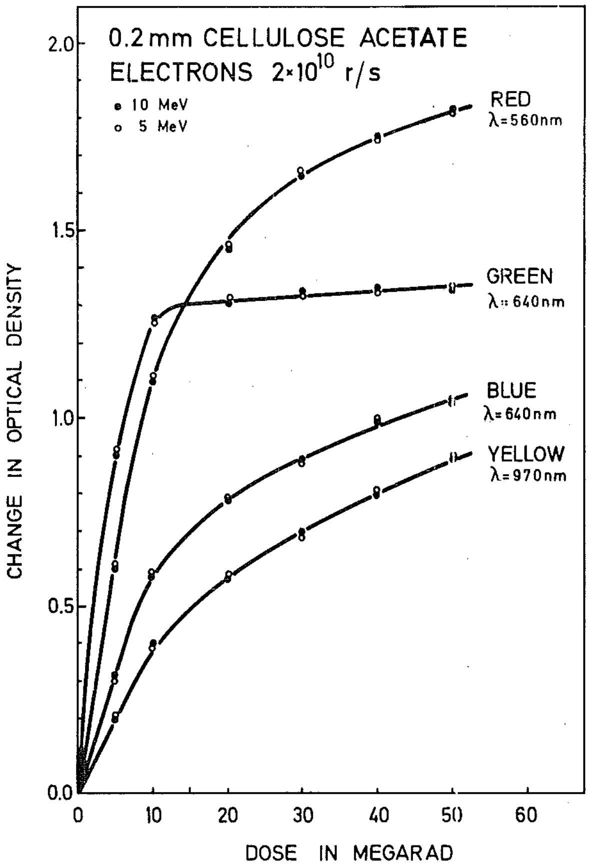

Fig. 15:

Dose response of coloured cellulose acetate irradiated with electrons of different energy

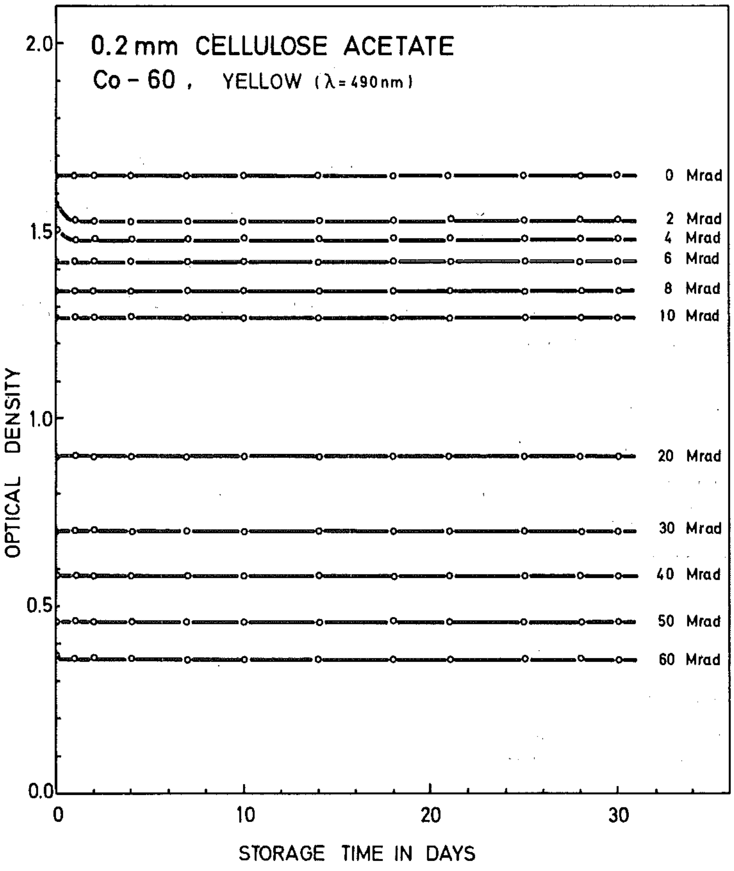

Fig. 16:

Optical density vs.storage time for yellow cellulose acetate exposed to different doses of γ-rays

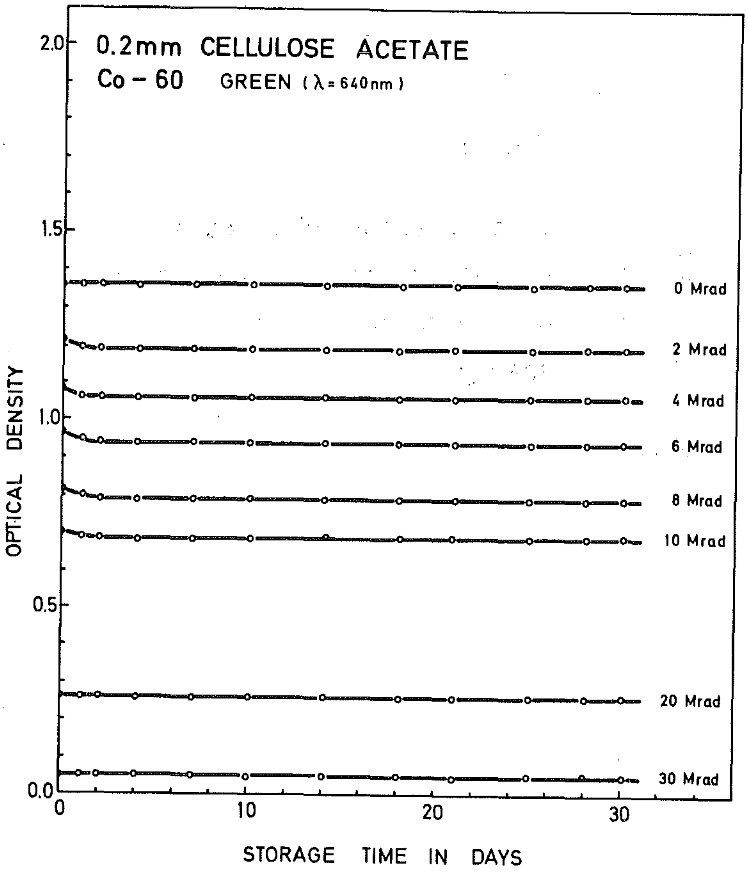

Fig. 17:

Optical density vs.storage time for green cellulose acetate exposed to different doses of γ-rays

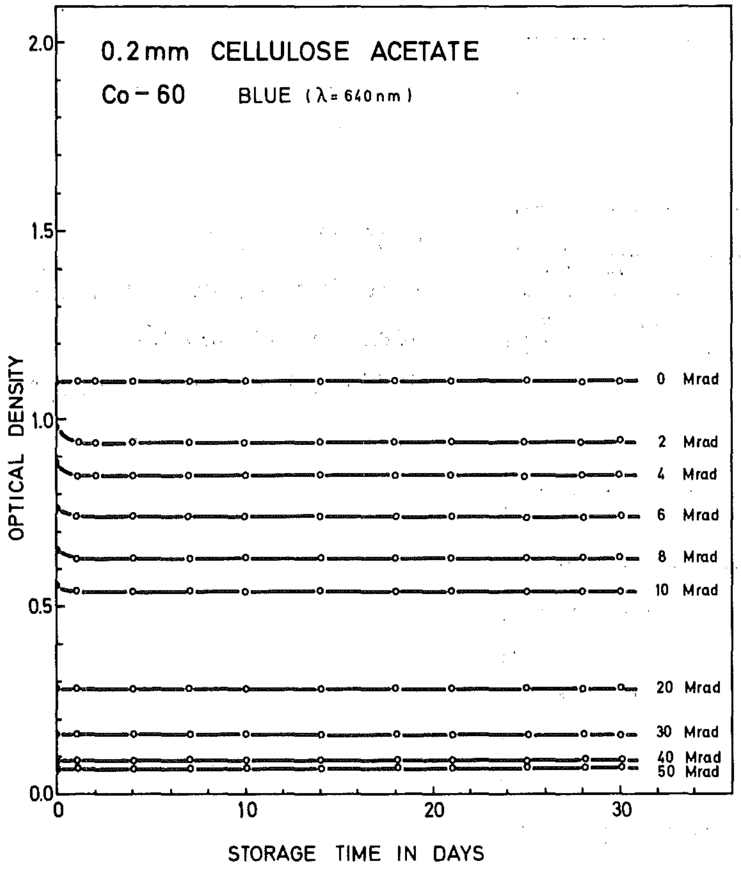

Fig. 18:

Optical density vs.storage time for blue cellulose acetate exposed to different doses of γ-rays

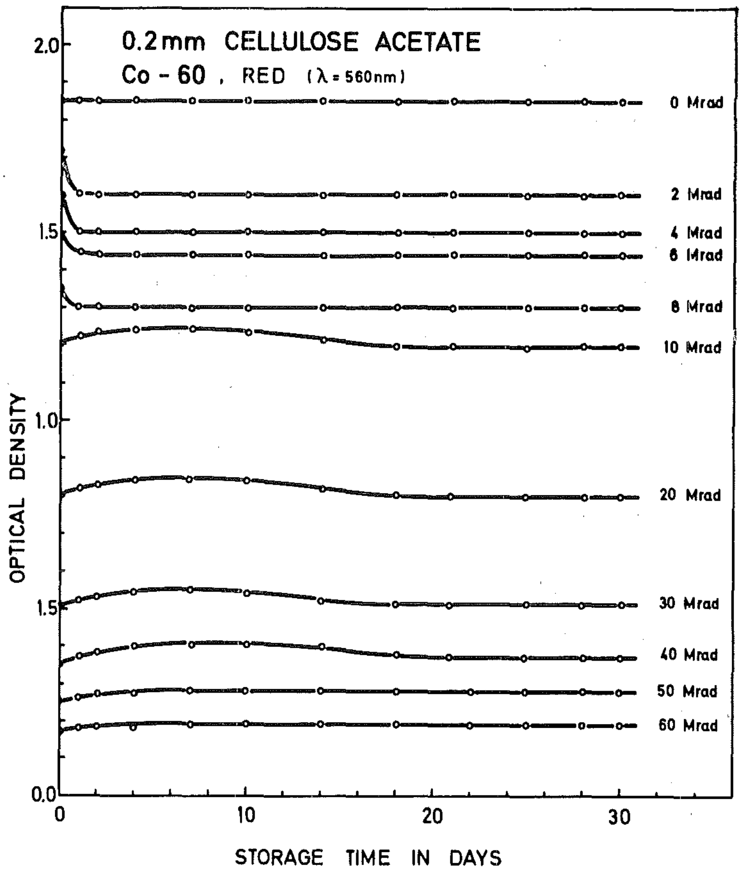

Fig. 19:

Optical density vs.storage time for red cellulose acetate exposed to different doses of γ-rays

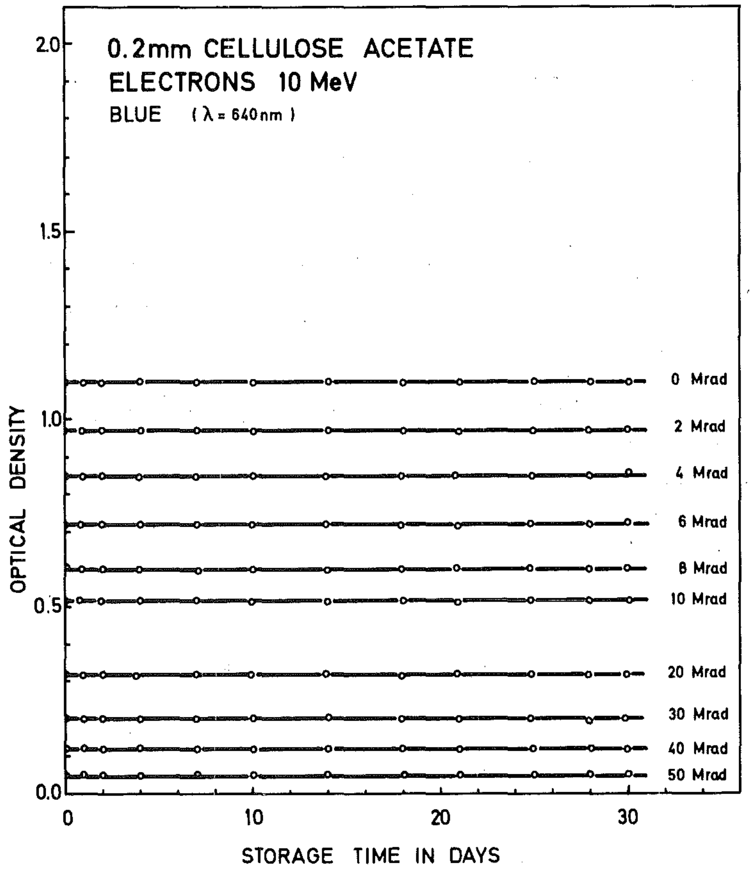

Fig. 20:

Optical density vs.storage time for blue cellulose acetate exposed to different doses of electrons

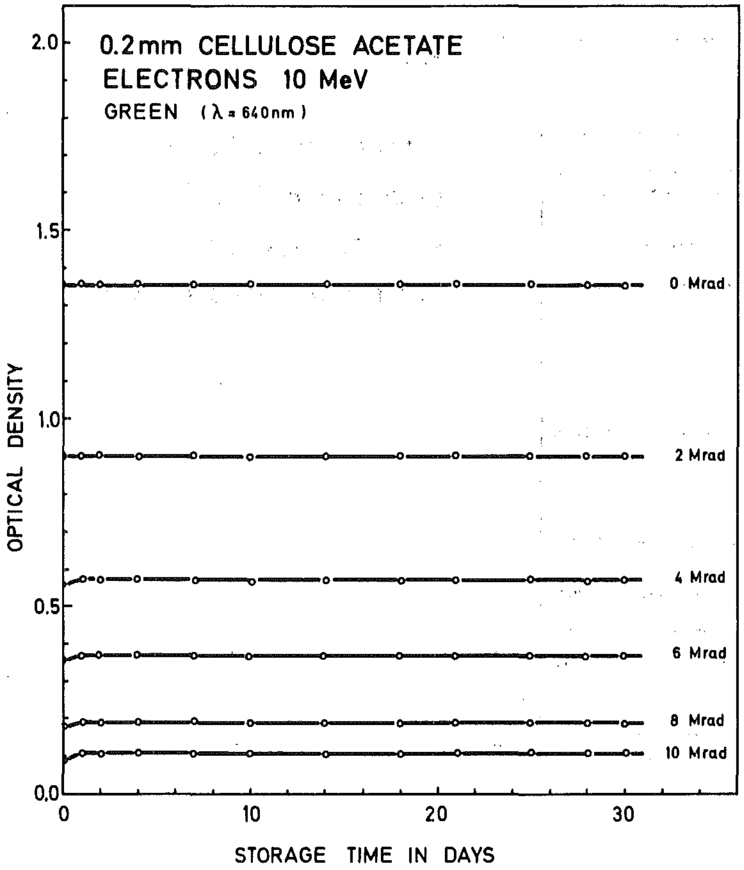

Fig. 21:

Optical density vs.storage time for green cellulose acetate exposed to different doses of electrons

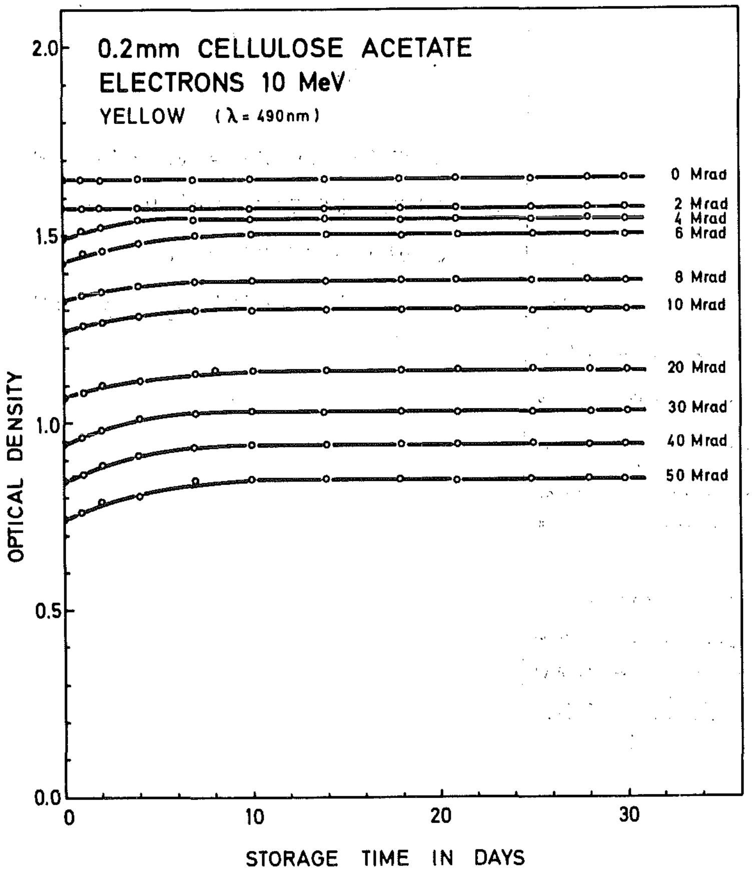

Fig. 22:

Optical density vs.storage time for yellow cellulose acetate exposed to different doses of electrons

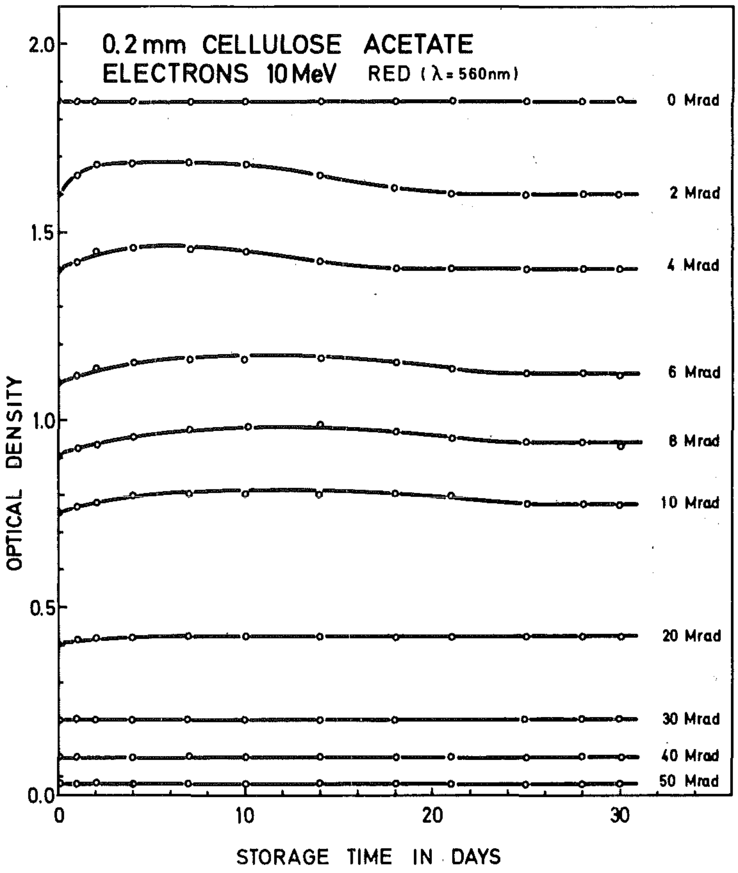

Fig. 23:

Optical density vs.storage time for red cellulose acetate exposed to different doses of electrons

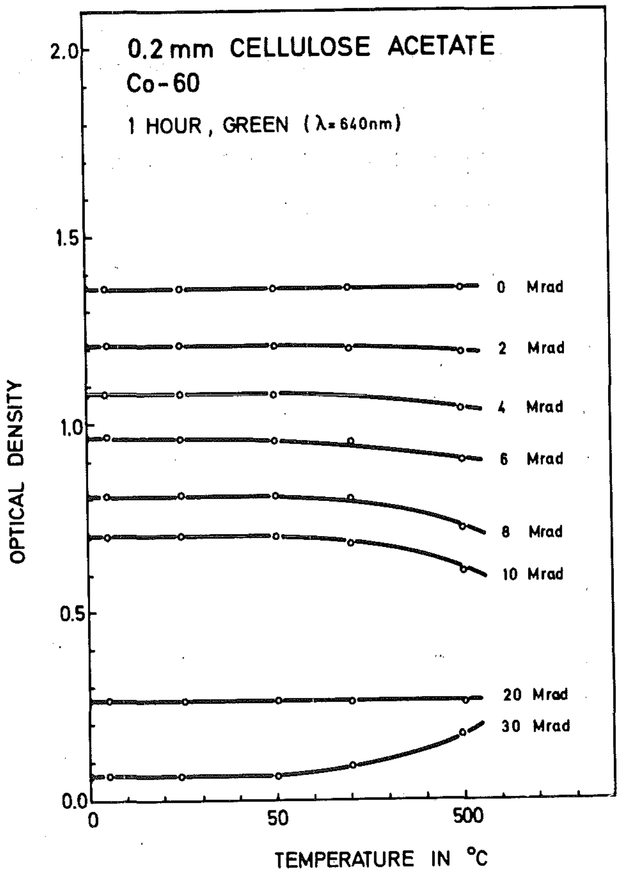

Fig. 24:

Optical density vs.temperature for green cellulose acetate exposed to different doses of γ-rays and stored for one hour

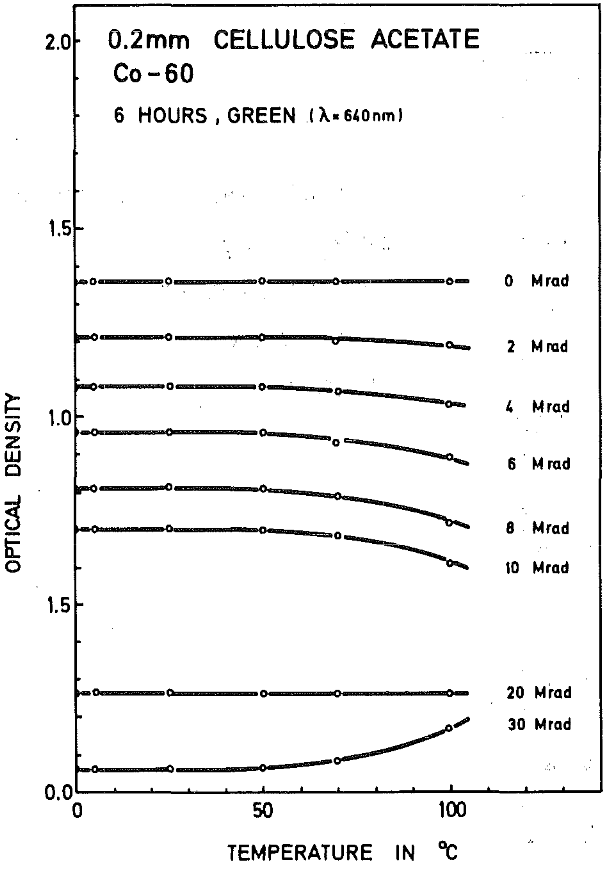

Fig. 25:

Optical density vs.temperature for green cellulose acetate exposed to different doses of γ-rays and stored for 6 hours

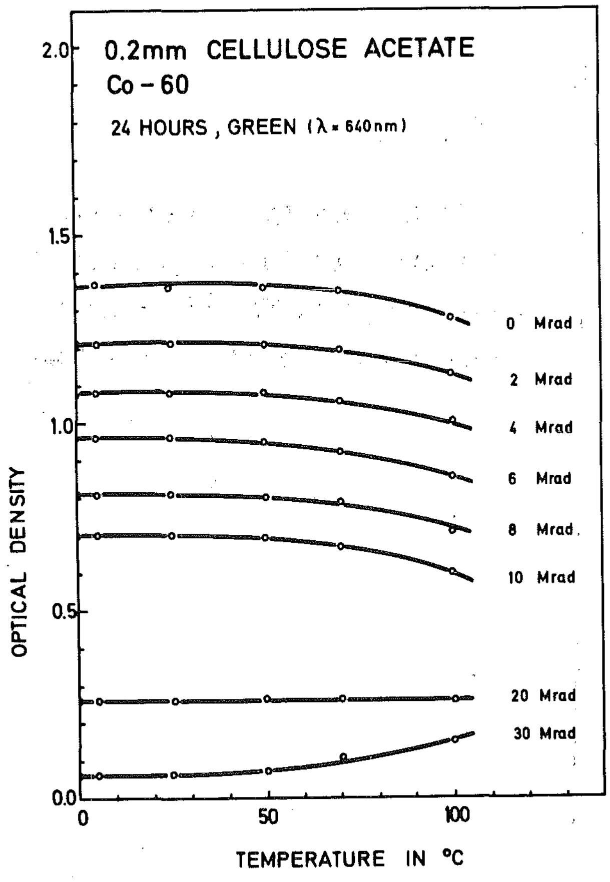

Fig. 26:

Optical density vs. temperature for green cellulose acetate exposed to different doses of γ-rays and stored for 24 hours

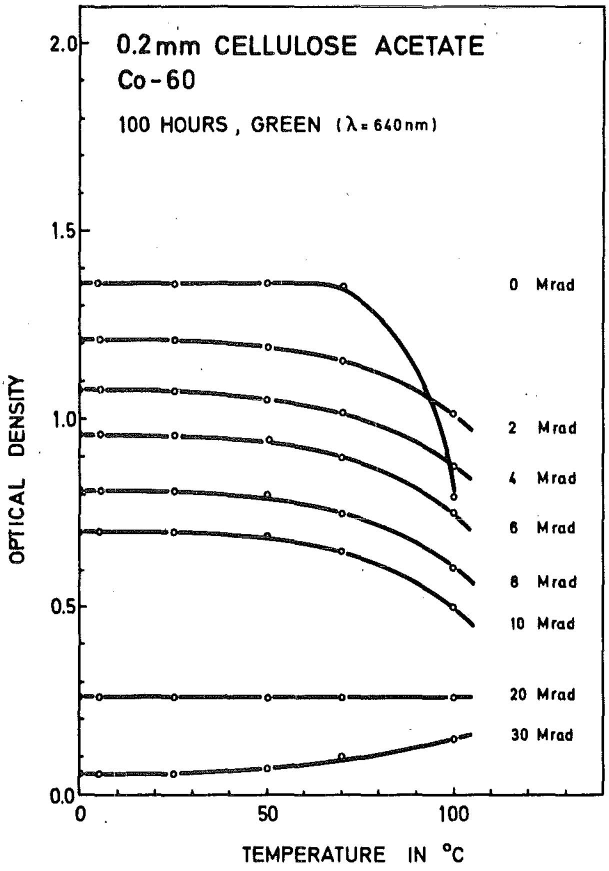

Fig. 27:

Optical density vs.temperature for green cellulose acetate exposed to different doses of γ-rays and stored for 100 hours

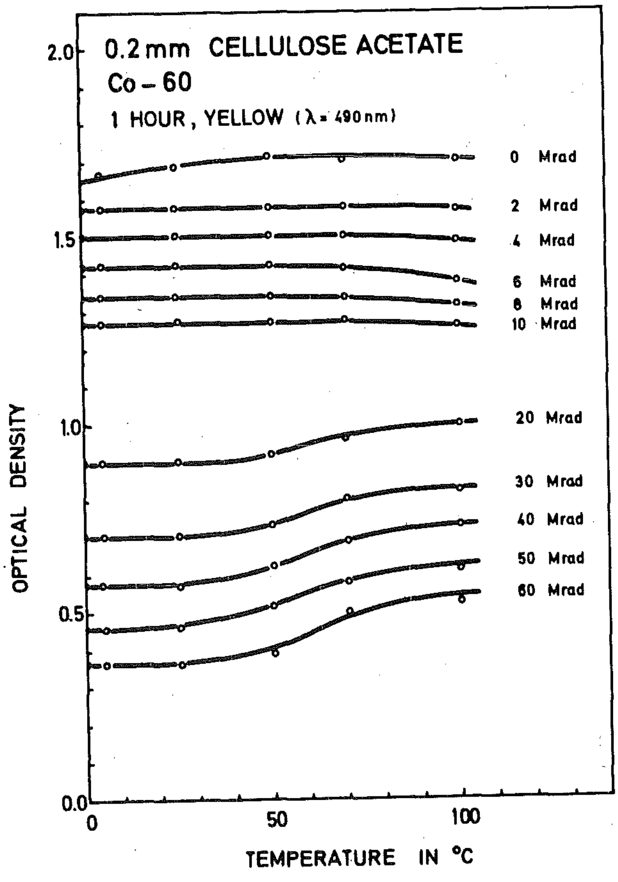

Fig. 28:

Optical density vs.temperature for yellow cellulose acetate exposed to different doses of γ-rays and stored for one hour

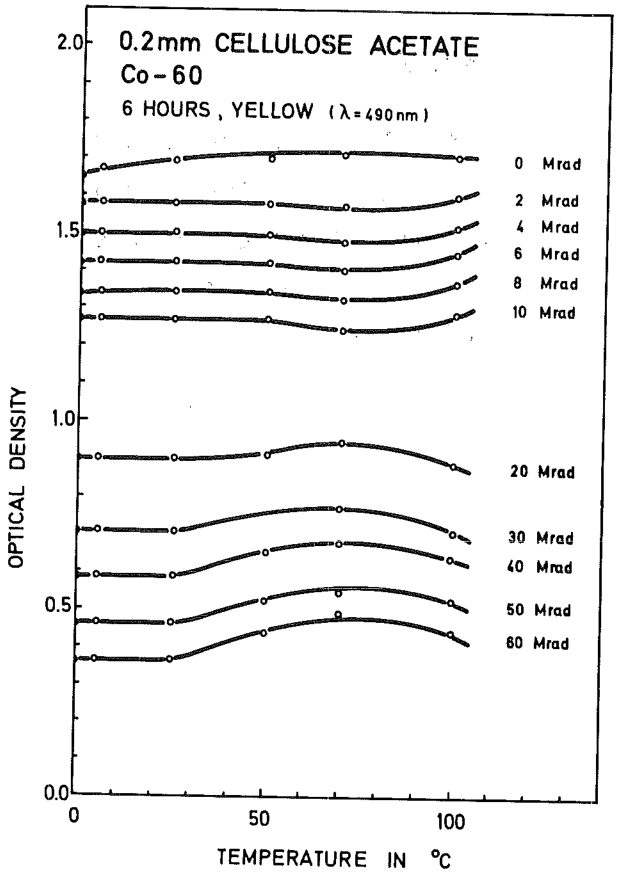

Fig. 29:

Optical density vs.temperature for yellow cellulose acetate exposed to different doses of γ-rays and stored for 6 hours

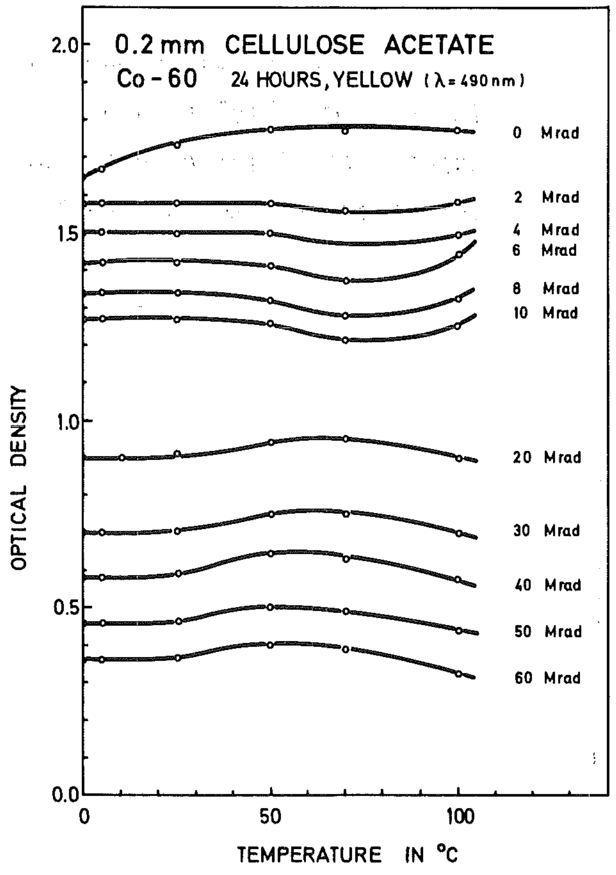

Fig. 30:

Optical density vs.temperature for yellow cellulose acetate exposed to different doses of γ-rays for 24 hours

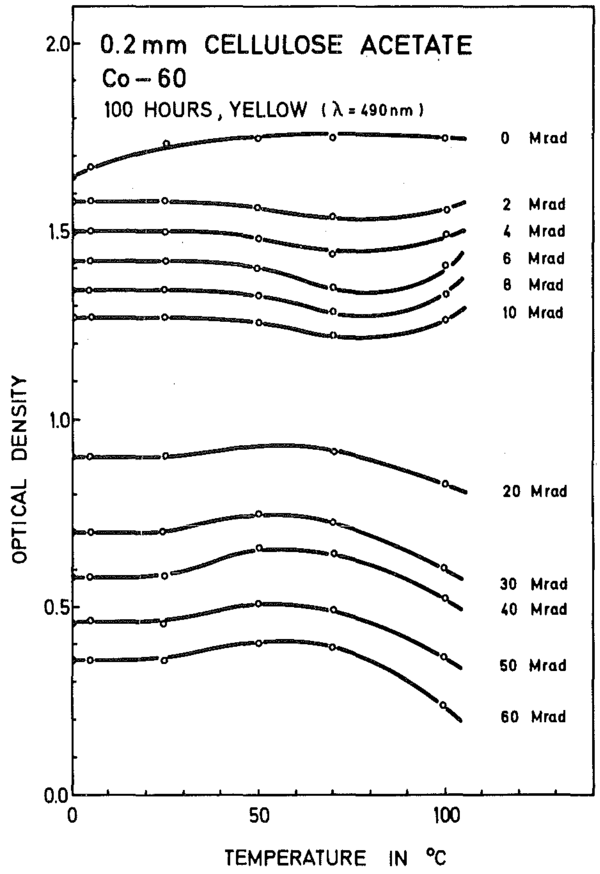

Fig. 31:

Optical density vs.temperature for yellow cellulose acetate exposed to different doses of γ-rays and stored for 100 hours

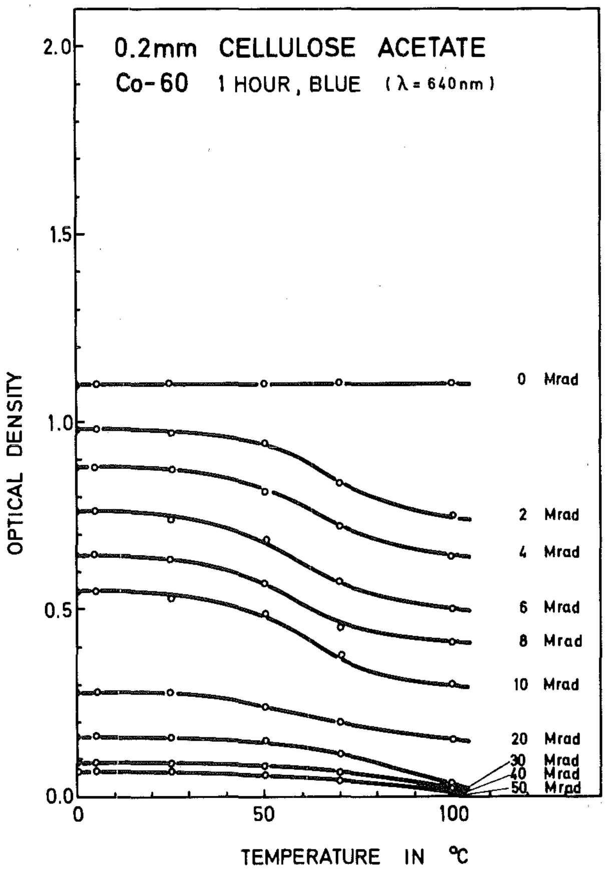

Fig. 32:

Optical density vs.temperature for blue cellulose acetate exposed to different doses of γ-rays and stored for one hour

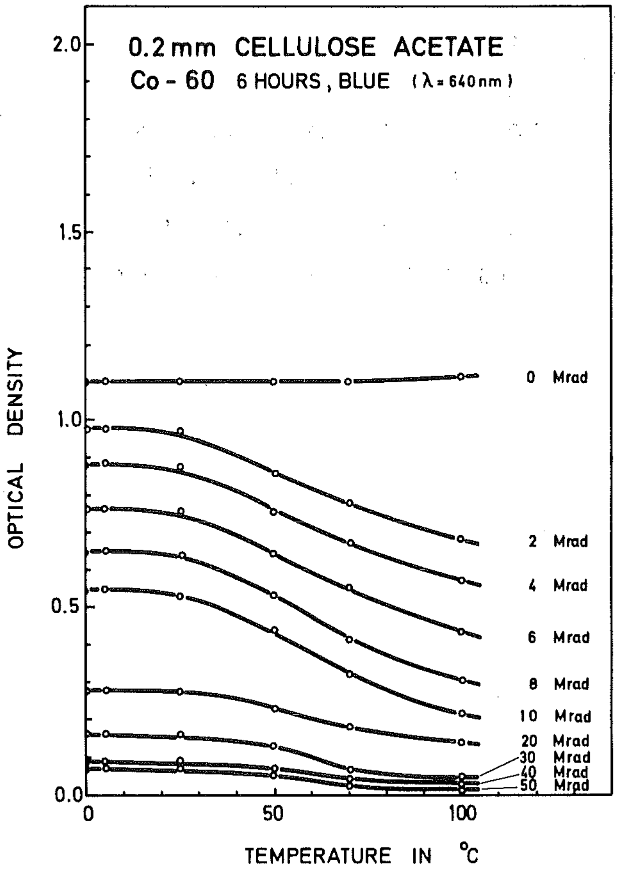

Fig. 33:

Optical density vs.temperature for blue cellulose acetate exposed to different doses of γ-rays and stored for 6 hours

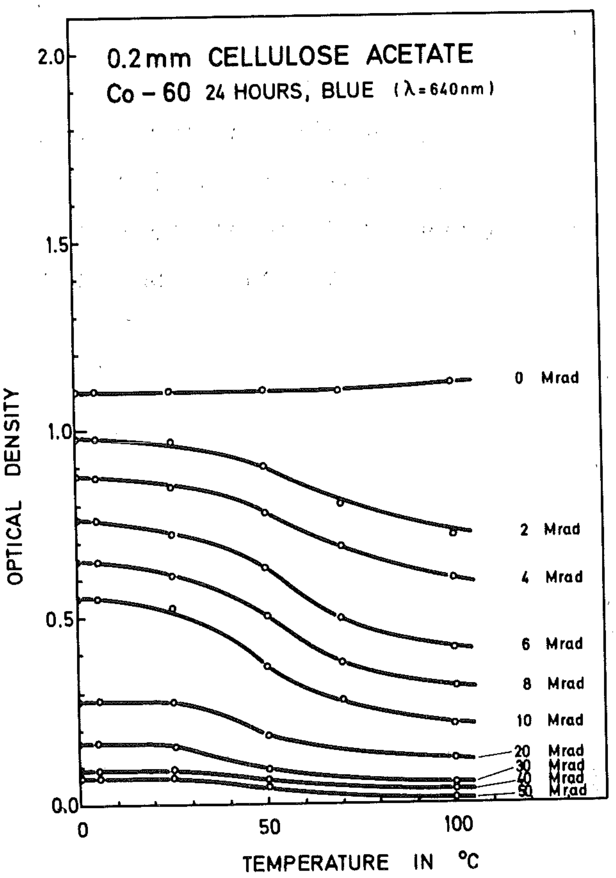

Fig. 34:

Optical density vs.temperature for blue cellulose acetate exposed to different doses of γ-rays and stored for 24 hours

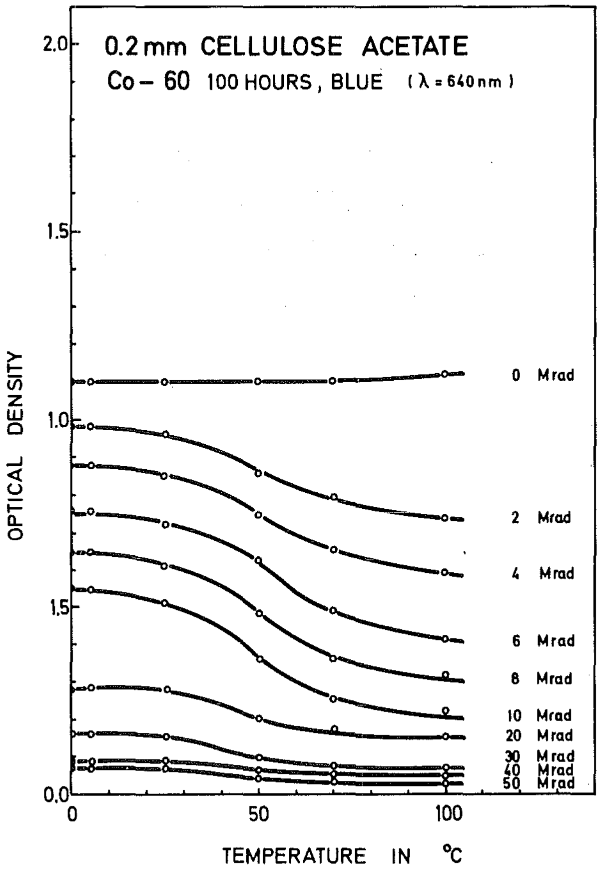

Fig. 35:

Optical density vs.temperature for blue cellulose acetate exposed to different doses of γ-rays and stored for 100 hours

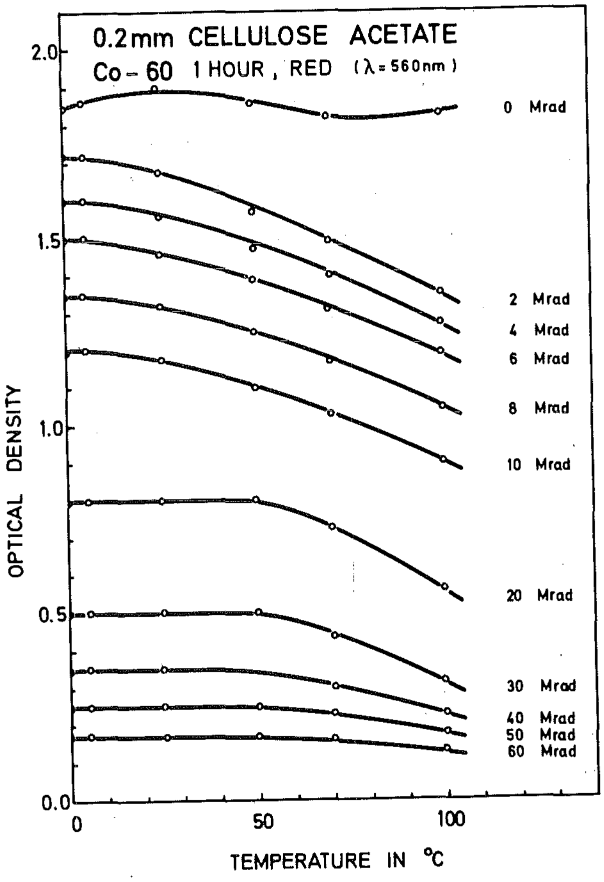

Fig. 36:

Optical density vs.temperature for red cellulose acetate exposed to different doses of γ-rays and stored for one hour

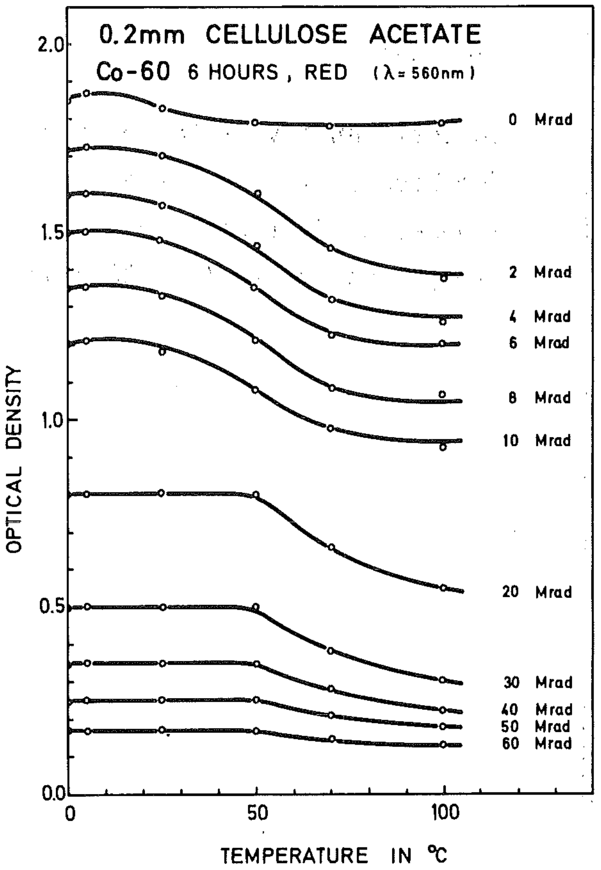

Fig. 37:

Optical density vs.temperature for red cellulose acetate exposed to different doses of γ-rays and stored for 6 hours

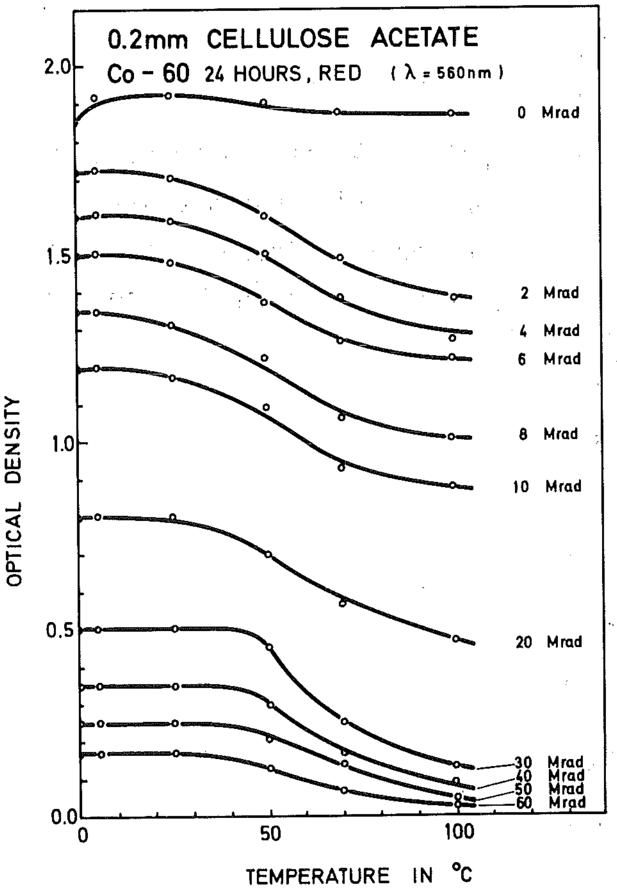

Fig. 38:

Optical density vs.temperature for red cellulose acetate exposed to different doses of γ-rays and stored for 24 hours

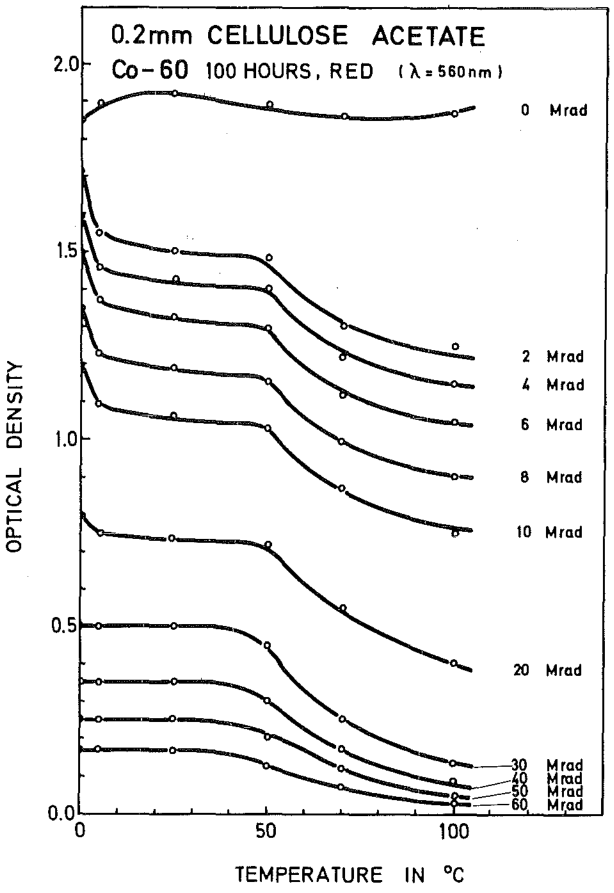

Fig. 39:

Optical density vs.temperature for red cellulose acetate exposed to different doses of γ-rays and stored for 100 hours

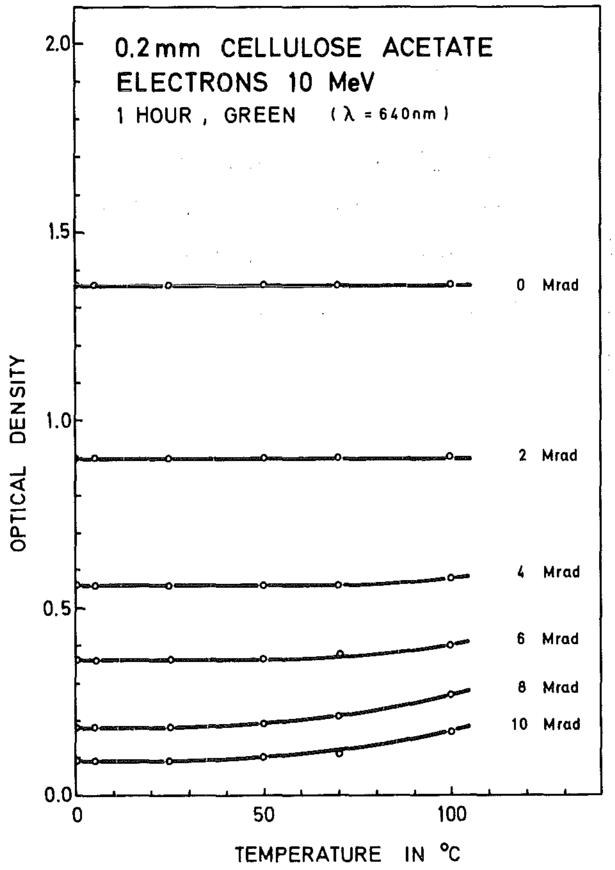

Fig. 40:

Optical density vs.temperature for green cellulose acetate exposed to different doses of electrons and stored for one hour

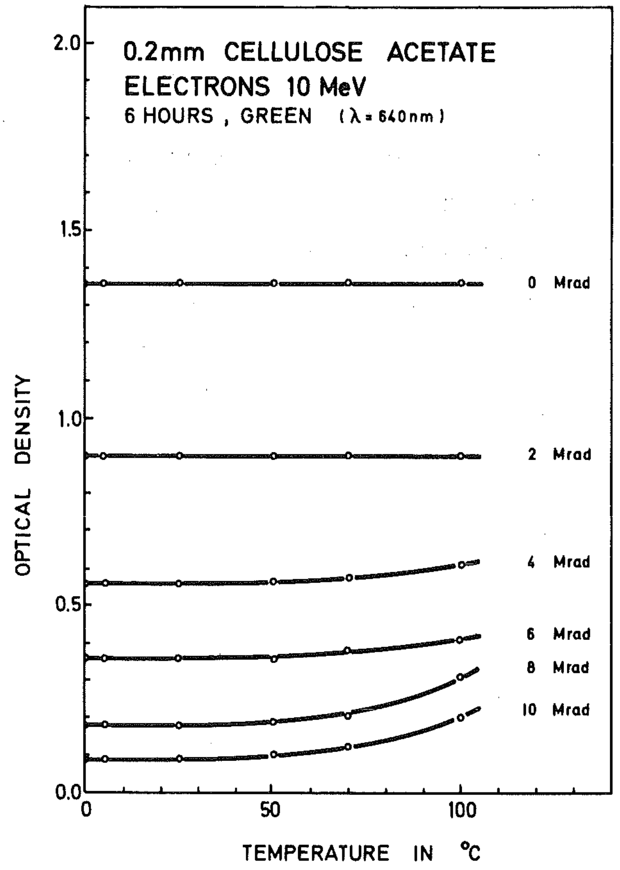

Fig. 41:

Optical density vs.temperature for green cellulose acetate exposed to different doses of electrons and stored for 6 hours

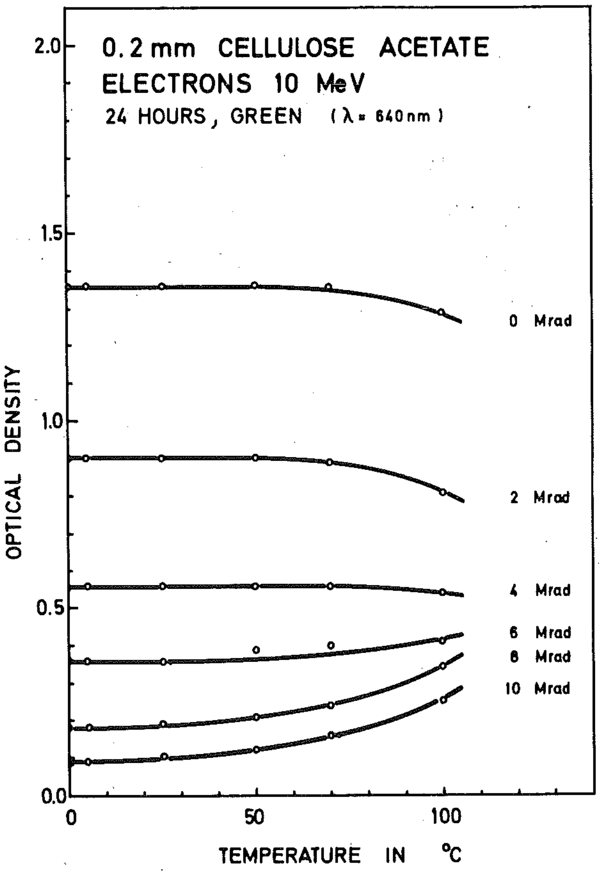

Fig. 42:

Optical density vs.temperature for green cellulose acetate exposed to different doses of electrons and stored for 24 hours

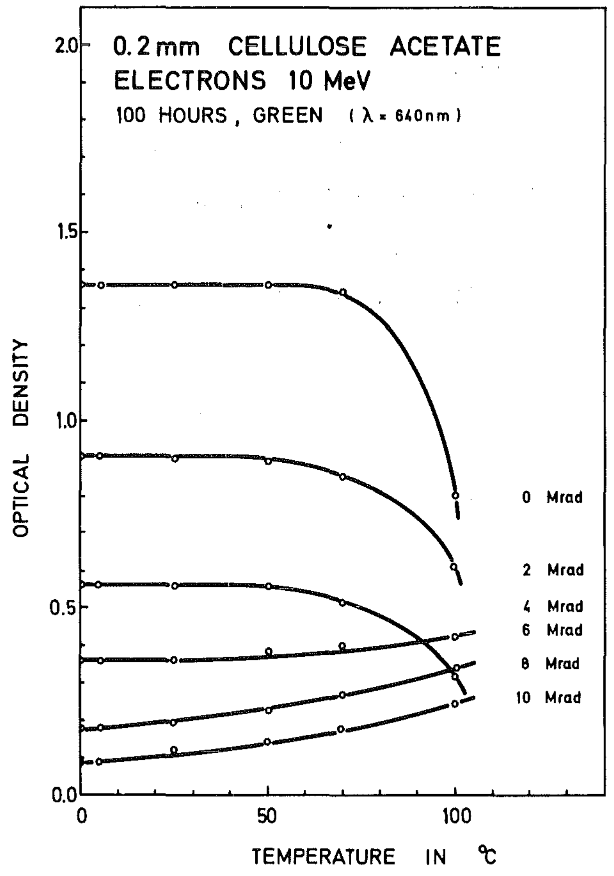

Fig. 43:

Optical density vs.temperature for green cellulose acetate exposed to different doses of electrons and stored for 100 hours

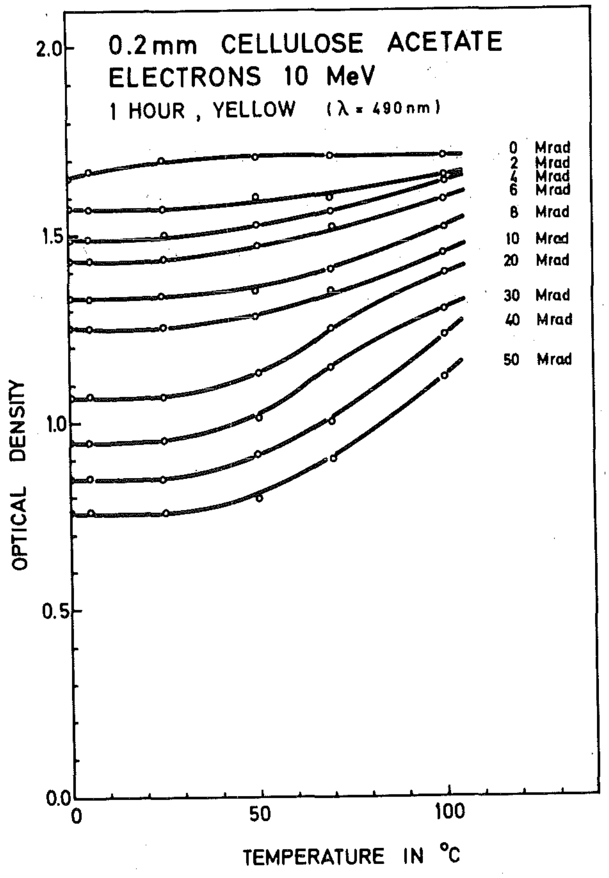

Fig. 44:

Optical density vs.temperature for yellow cellulose acetate exposed to different doses of electrons and stored for one hour

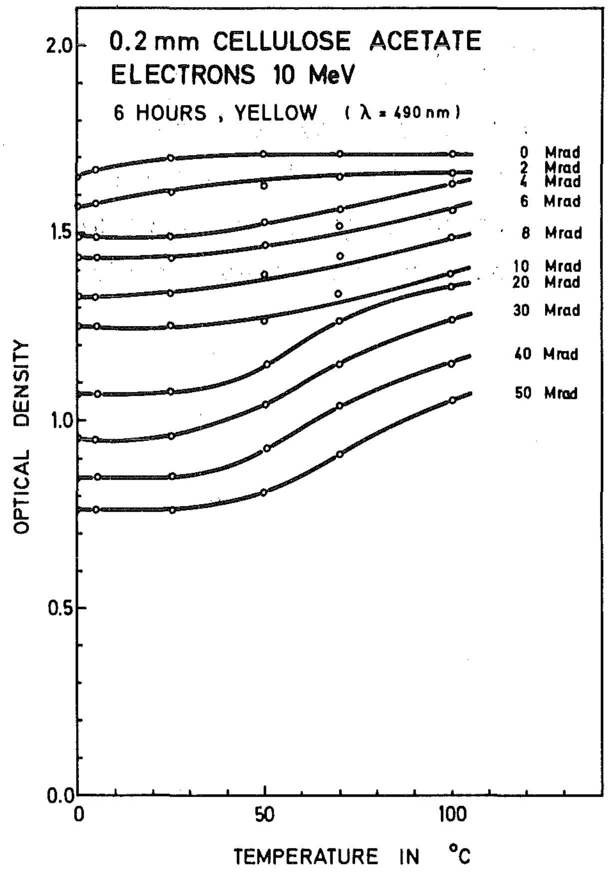

Fig. 45:

Optical density vs.temperature for yellow cellulose acetate exposed to different doses of electrons and stored for 6 hours

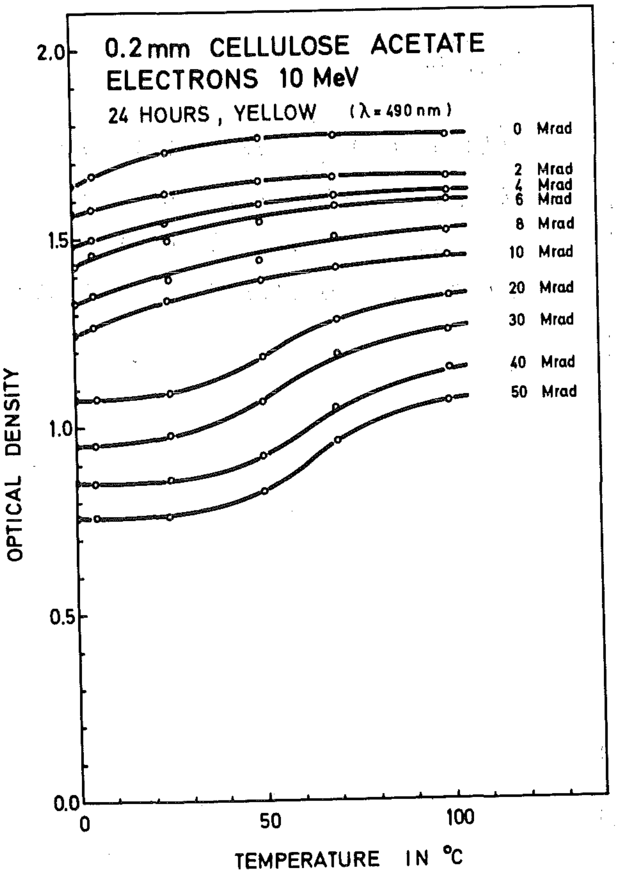

Fig. 46:

Optical density vs.temperature for yellow cellulose acetate exposed to different doses of electrons and stored for 24 hours

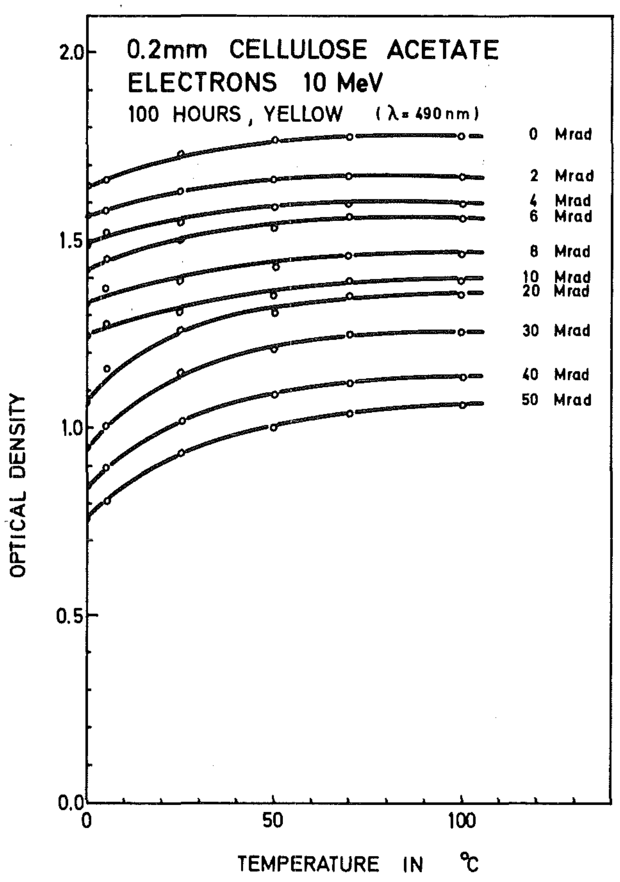

Fig. 47:

Optical density vs.temperature for yellow cellulose acetate exposed to different doses of electrons and stored for 100 hours

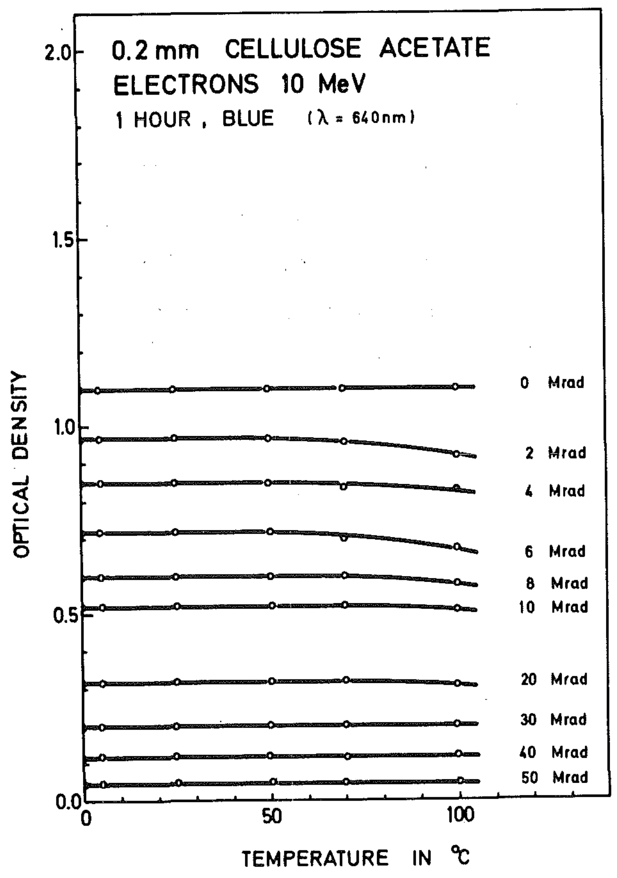

Fig. 48:

Optical density vs.temperature for blue cellulose acetate exposed to different doses of electrons and stored for one hour

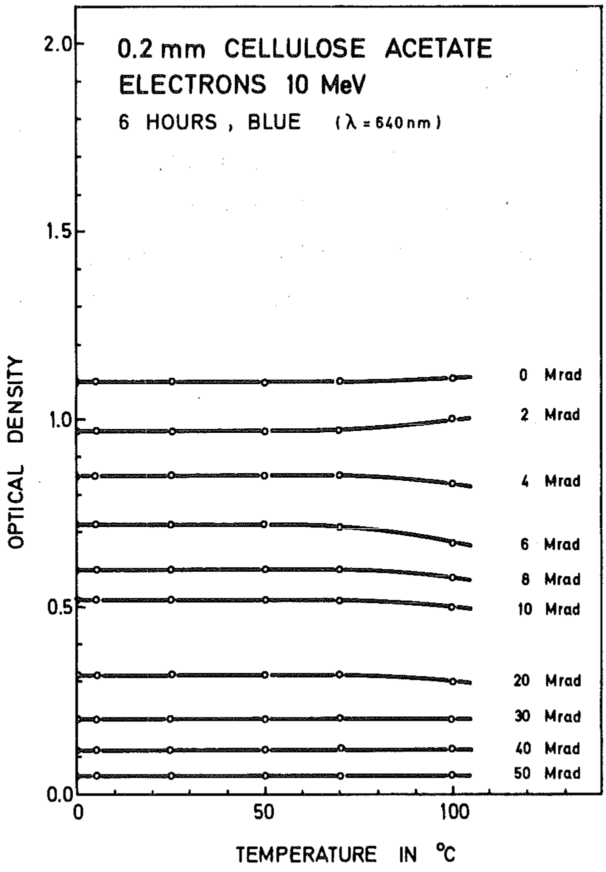

Fig. 49:

Optical density vs.temperature for blue cellulose acetate exposed to different doses of electrons and stored for 6 hours

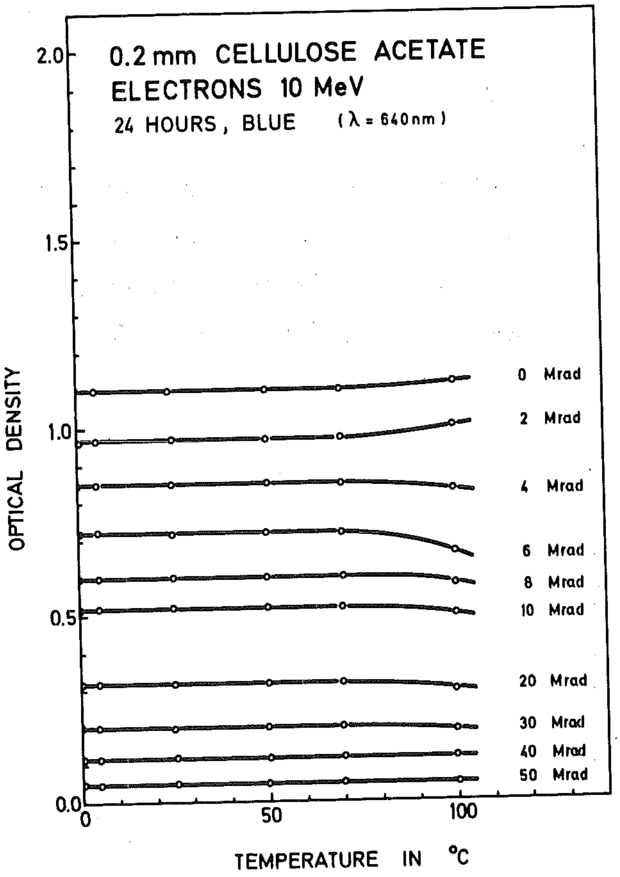

Fig. 50:

Optical density vs.temperature for blue cellulose acetate exposed to different doses of electrons and stored for 24 hours

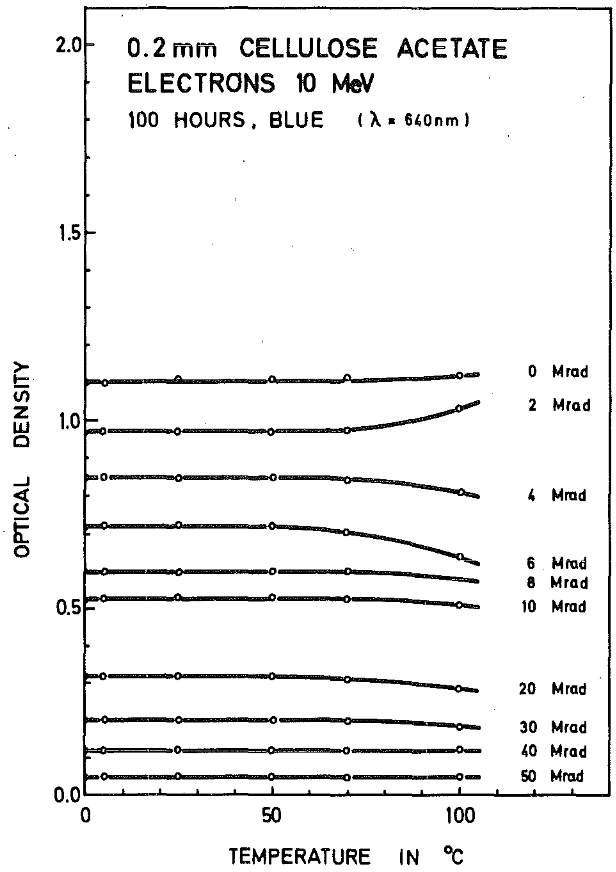

Fig. 51:

Optical density vs.temperature for blue cellulose acetate exposed to different doses of electrons and stored for 100 hours

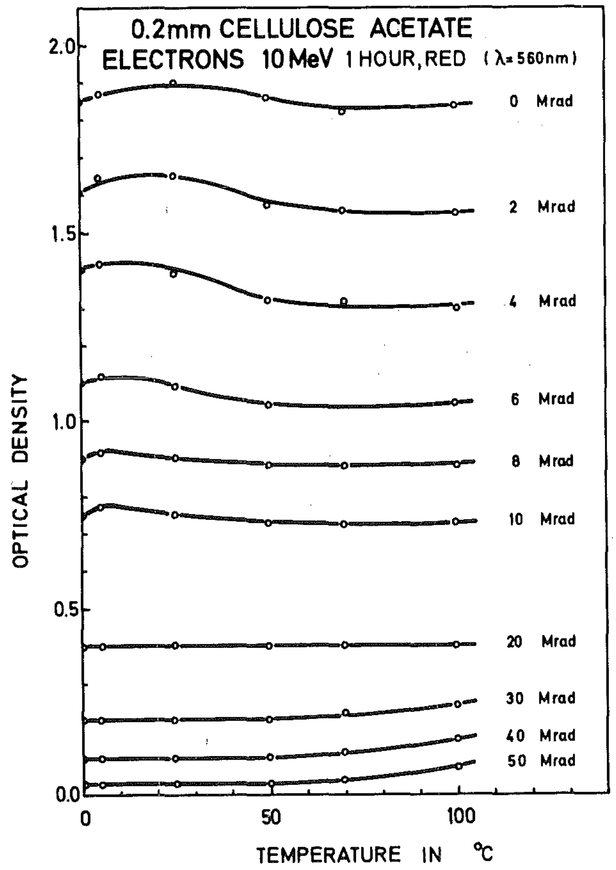

Fig. 52:

Optical density vs. temperature for red cellulose acetate exposed to different doses of electrons and stored for one hour

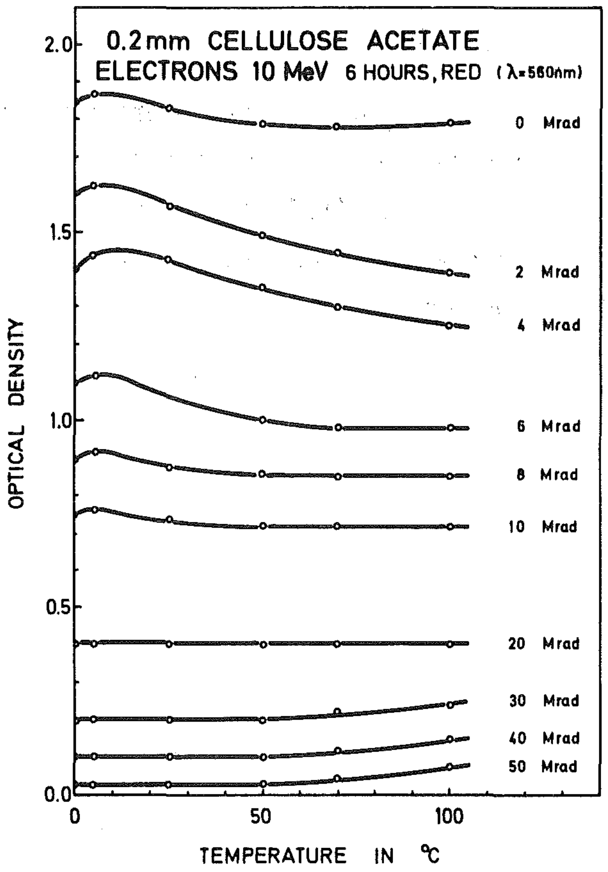

Fig. 53:

Optical density vs.temperature for red cellulose acetate exposed to different doses of electrons and stored for 6 hours

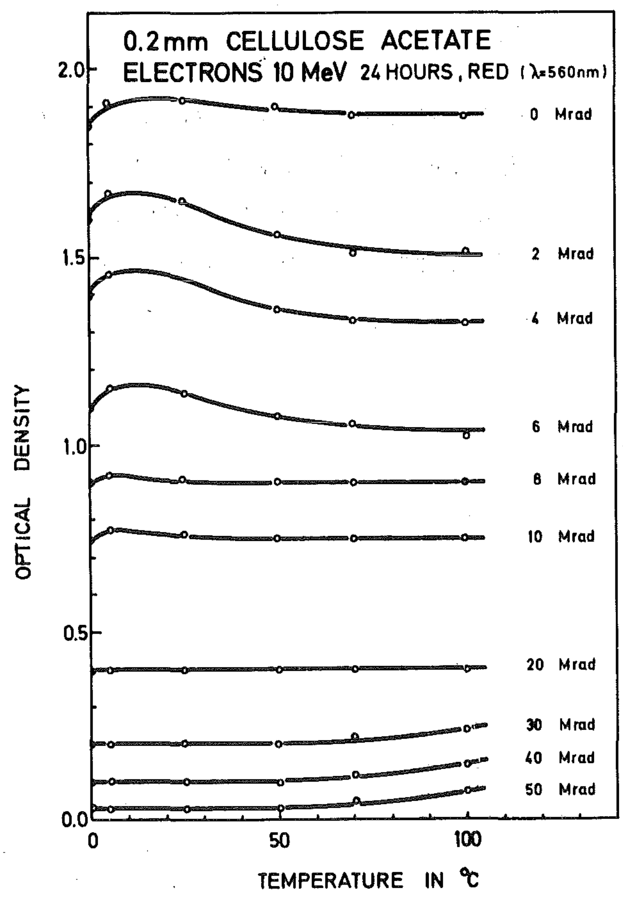

Fig. 54:

Optical density vs.temperature for red cellulose acetate exposed to different doese of electrons and stored for 24 hours

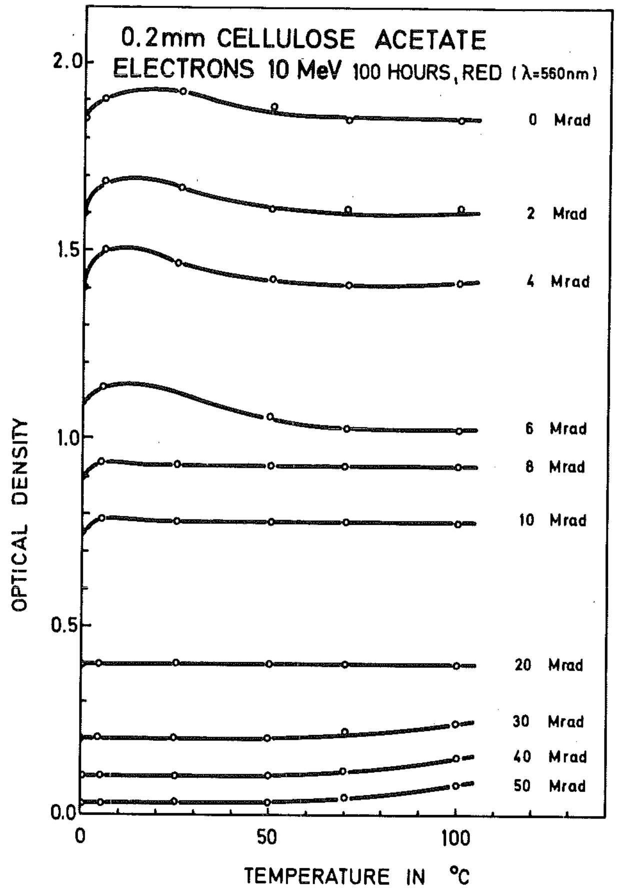

Fig. 55:

Optical density vs.temperature for red cellulose acetate exposed to different doses of electrons and stored for 100 hours

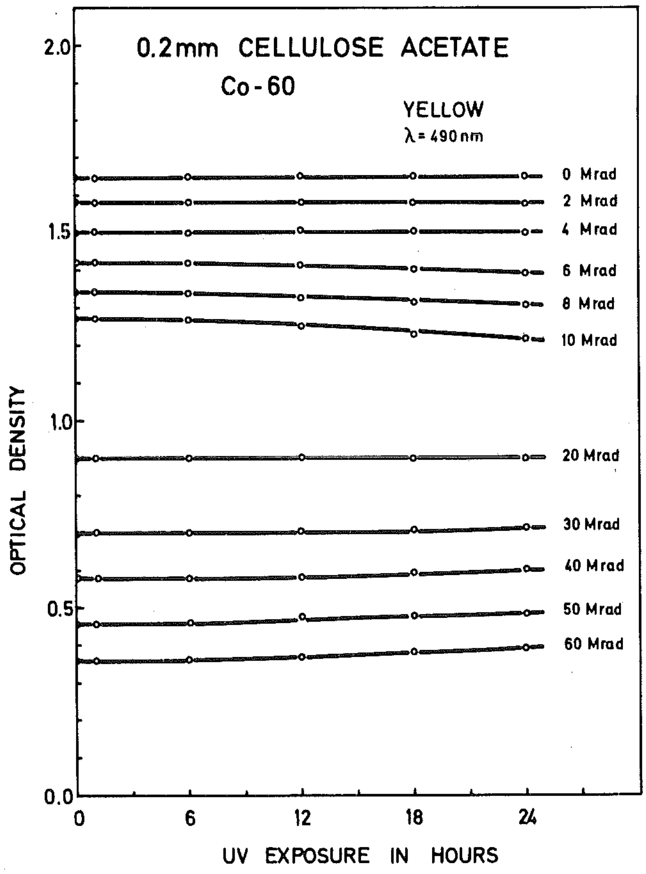

Fig. 56:

Optical density vs.UV exposure for yellow cellulose acetate exposed to different doses of γ-rays

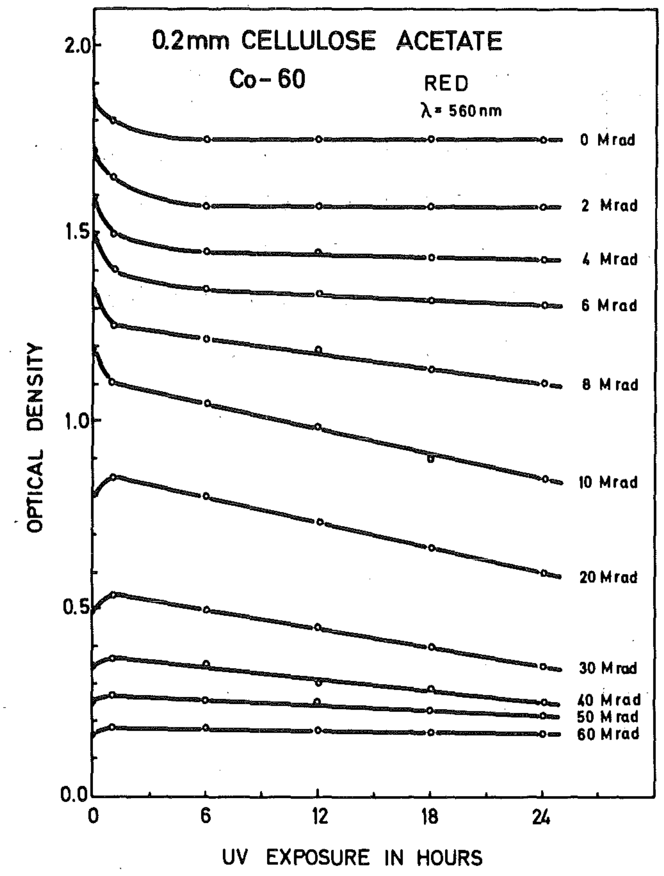

Fig. 57:

Optical density vs.UV exposure for red cellulose acetate exposed to different doses of γ-rays

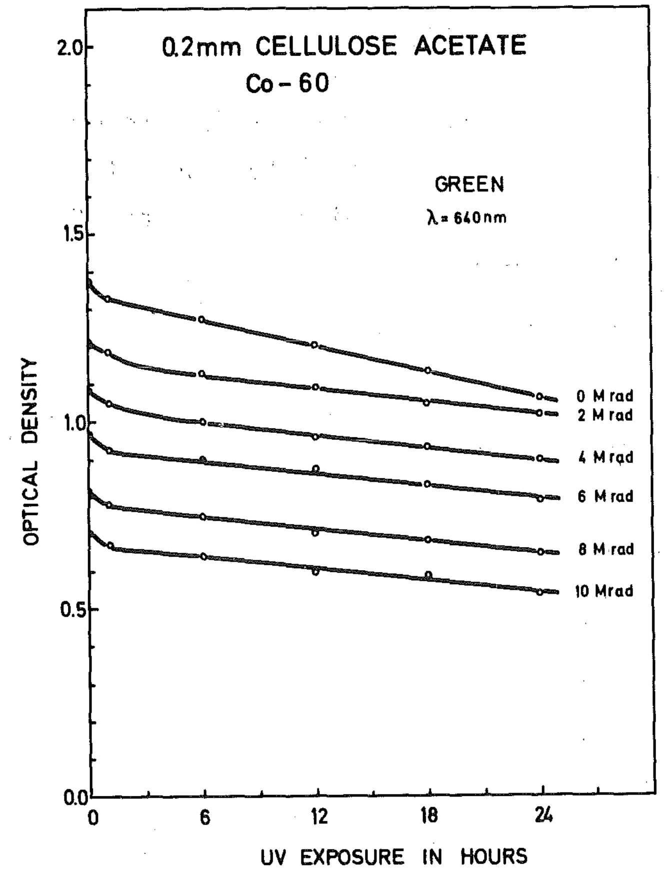

Fig. 58:

Optical density vs.UV exposure for green cellulose acetate exposed to different doses of γ-rays

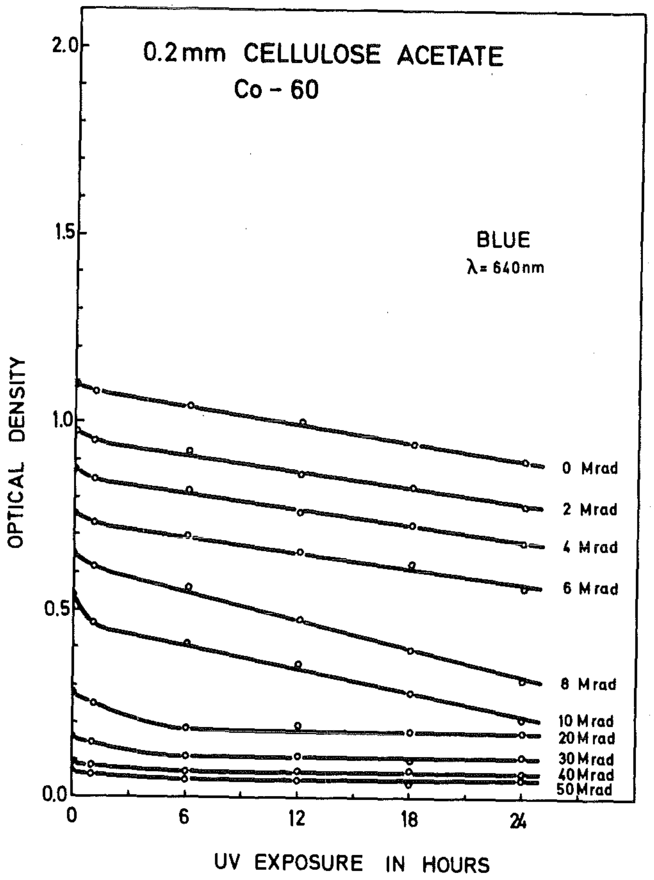

Fig. 59:

Optical density vs.UV exposure for blue cellulose acetate exposed to different doses of γ-rays

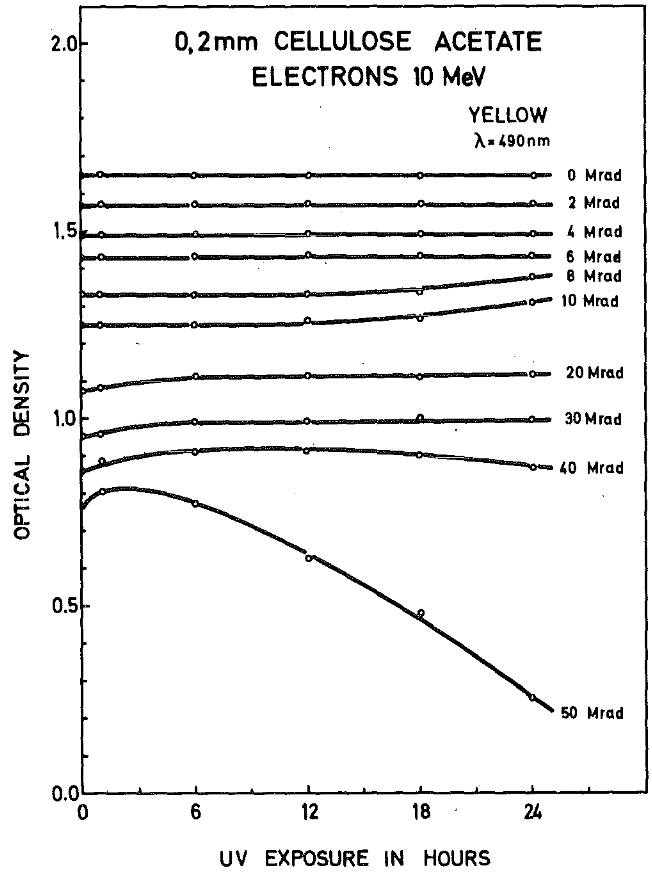

Fig. 60:

Optical density vs.UV exposure for yellow cellulose acetate exposed to different doses of electrons

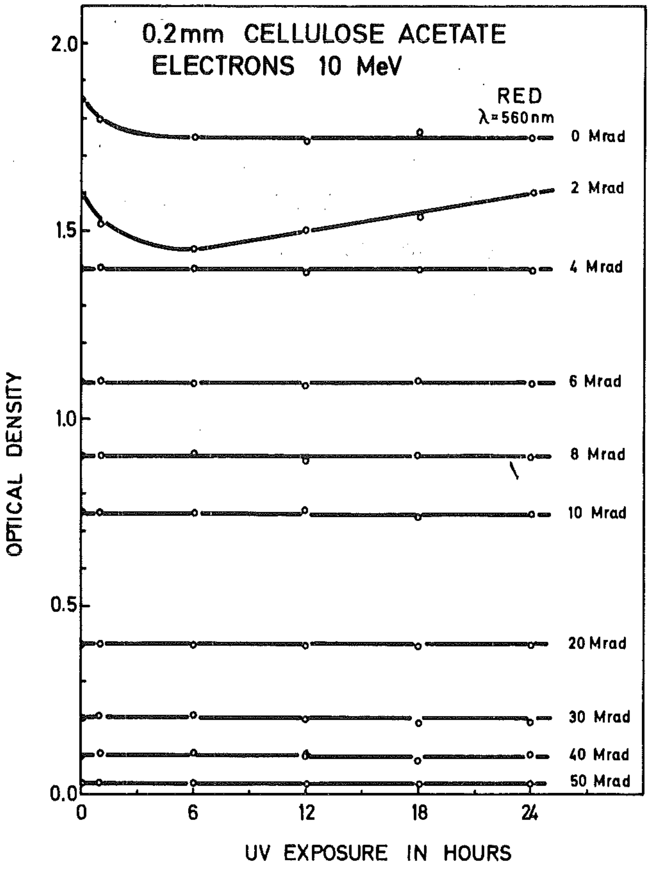

Fig. 61:

Optical density vs.UV exposure for red cellulose acetate exposed to different doses of electrons

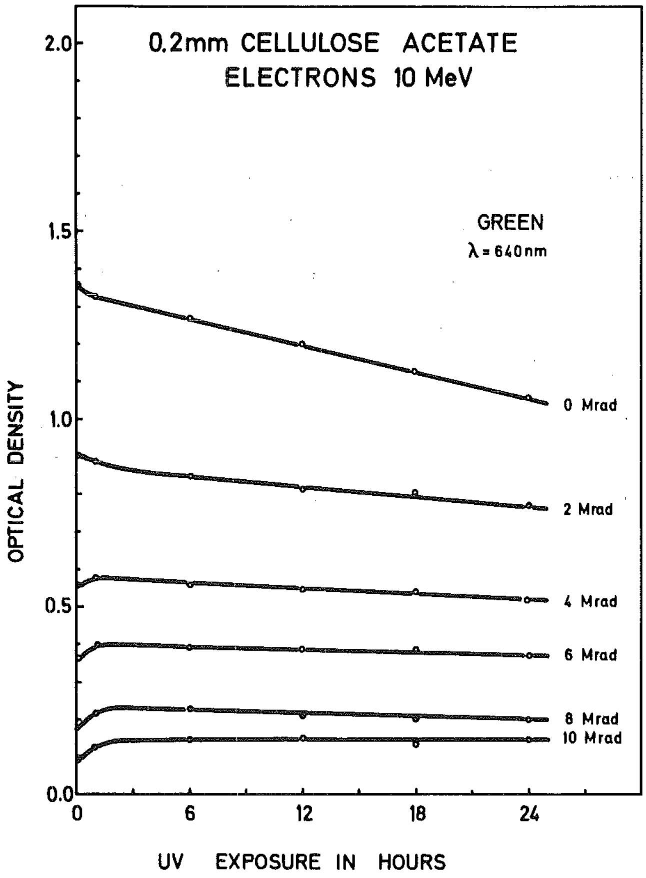

Fig. 62:

Optical density vs.UV exposure for green cellulose acetate exposed to different doses of electrons

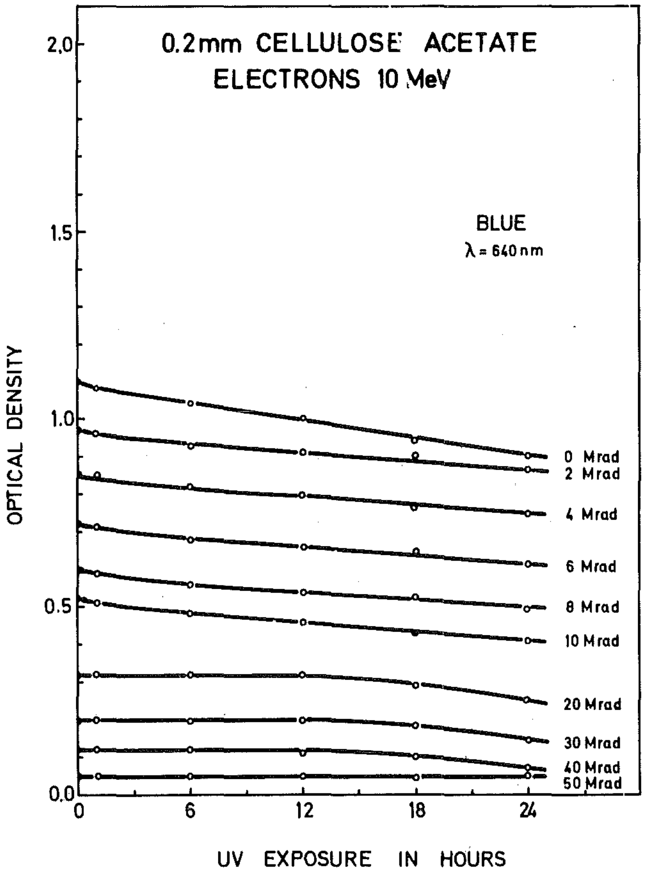

Fig. 63:

Optical density vs.UV exposure for blue cellulose acetate exposed to different doses of electrons

Figure captions

Fig. 1 Change in optical density of coloured cellulose acetate irradiated with γ-rays

Fig. 2 Change in % transmission of coloured cellulose acetate irradiated with γ-rays

Fig. 3 Change in optical density of coloured cellulose acetate irradiated with electrons

Fig. 4 Change in % transmission of coloured cellulose acetate irradiated with electrons

Fig. 5 Dose response of yellow cellulose acetate irradiated with γ-rays

Fig. 6 Dose response of red cellulose acetate irradiated with γ-rays

Fig. 7 Dose response of green cellulose acetate irradiated with γ-rays

Fig. 8 Dose response of blue cellulose acetate irradiated with γ-rays

Fig. 9 Dose response of yellow cellulose acetate irradiated with electrons

Fig. 10 Dose response of green cellulose acetate irradiated with electrons

Fig. 11 Dose response of red cellulose acetate irradiated with electrons

Fig. 12 Dose response of blue cellulose acetate irradiated with electrons

Fig. 13 Dose response of coloured cellulose acetate irradiated with γ-rays of different dose rate

Fig. 14 Dose response of coloured cellulose acetate irradiated with electrons of different dose rate

Fig. 15 Dose response of coloured cellulose acetate irradiated with electrons of different energy

Fig. 16 Optical density vs.storage time for yellow cellulose acetate exposed to different doses of γ-rays

Fig. 17 Optical density vs.storage time for green cellulose acetate exposed to different doses of γ-rays

Fig. 18 Optical density vs.storage time for blue cellulose acetate exposed to different doses of γ-rays

Fig. 19 Optical density vs.storage time for red cellulose acetate exposed to different doses of γ-rays

Fig. 20 Optical density vs.storage time for blue cellulose acetate exposed to different doses of electrons

Fig. 21 Optical density vs.storage time for green cellulose acetate exposed to different doses of electrons

Fig. 22 Optical density vs.storage time for yellow cellulose acetate exposed to different doses of electrons

Fig. 23 Optical density vs.storage time for red cellulose acetate exposed to different doses of electrons

Fig. 24 Optical density vs.temperature for green cellulose acetate exposed to different doses of γ-rays and stored for one hour

Fig. 25 Optical density vs.temperature for green cellulose acetate exposed to different doses of γ-rays and stored for 6 hours

Fig. 26 Optical density vs.temperature for green cellulose acetate exposed to different doses of γ-rays and stored for 24 hours

Fig. 27 Optical density vs.temperature for green cellulose acetate exposed to different doses of γ-rays and stored for 100 hours

Fig. 28 Optical density vs.temperature for yellow cellulose acetate exposed to different doses of γ-rays and stored for one hour

Fig. 29 Optical density vs.temperature for yellow cellulose acetate exposed to different doses of γ-rays and stored for 6 hours

Fig. 30 Optical density vs.temperature for yellow cellulose acetate exposed to different doses of γ-rays and stored for 24 hours

Fig. 31 Optical density vs.temperature for yellow cellulose acetate exposed to different doses of γ-rays and stored for 100 hours

Fig. 32 Optical density vs.temperature for blue cellulose acetate exposed to different doses of γ- rays and stored for one hour

Fig. 33 Optical density vs.temperature for blue cellulose acetate exposed to different doses of γ-rays and stored for 6 hours

Fig. 34 Optical density vs.temperature for blue cellulose acetate exposed to different doses of γ-rays and stored for 24 hours

Fig. 35 Optical density vs. temperature for blue cellulose acetate exposed to different doses of γ-rays and stored for 100 hours

Fig. 52	Optical density vs. temperature for red cellulose acetate exposed to different doses of electrons and stored for one hour
Fig. 53	Optical density vs. temperature for red cellulose acetate exposed to different doses of electrons and stored for 6 hours
Fig. 54	Optical density vs. temperature for red cellulose acetate exposed to different doses of electrons and stored for 24 hours
Fig. 55	Optical density vs. temperature for red cellulose acetate exposed to different doses of electrons and stored for 100 hours
Fig. 56	Optical density vs. UV exposure for yellow cellulose acetate exposed to different doses of γ-rays
Fig. 57	Optical density vs. UV exposure for red cellulose acetate exposed to different doses of γ-rays
Fig. 58	Optical density vs. UV exposure for green cellulose acetate exposed to different doses of γ-rays
Fig. 59	Optical density vs. UV exposure for blue cellulose acetate exposed to different doses of γ-rays
Fig. 60	Optical density vs. UV exposure for yellow cellulose acetate exposed to different doses of electrons
Fig. 61	Optical density vs. UV exposure for red cellulose acetate exposed to different doses of electrons
Fig. 62	Optical density vs. UV exposure for green cellulose acetate exposed to different doses of electrons
Fig. 63	Optical density vs. UV exposure for blue cellulose acetate exposed to different doses of electrons