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EFFECT OF PRE–GAMMA IRRADIATION ON CR–39*

N. E. IPE

Stanford Linear Accelerator Center
Stanford University, Stanford, California, 94305

and

P. L. ZIEMER

School of Health Sciences
Purdue University, West Lafayette, Indiana 47907

ABSTRACT

The effect of pre-gamma irradiation on track density, bulk etch rate and light absorbance in CR-39 was investigated. CR-39 detectors were irradiated with gamma doses over a range of 0 to 280 kGy (0 to 28 Mrad). The detectors were overlaid with 0.77 mm of polyethylene and exposed to a known fluence of ^{252}Cf spontaneous fission neutrons. All detectors were then etched in 6.25N sodium hydroxide solution at 70°C for 4 hours and track densities were determined with an optical microscope. The bulk etch rates were determined by measuring the change in thickness before and after etching. The net track density decreased and the bulk etch rate increased non-linearly with increasing gamma doses. UV spectrophotometric measurements showed significant changes in light absorbance for gamma irradiated CR-39.

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INTRODUCTION

CR-39 is a polymeric nuclear track detector which is currently being used for fast neutron detection. During use nuclear track detectors may sometimes be exposed to high intensity gamma radiation fields. Such gamma exposure can induce changes in the bulk detector material and in the latent damage tracks, if the damage tracks were induced in the material before or during gamma irradiation. Correction of any induced detector effect requires a knowledge of the nature of this effect. At the same time a knowledge of this effect may offer the prospects for measuring gamma dose. Some aspects of the effect of gamma radiation on CR-39 have been reported.(1,2,3) This paper reports the effect of pre-gamma irradiation on neutron induced track density and bulk etch rate in CR-39.

Ultraviolet light absorbance studies are powerful techniques for the measurement of gamma dose and the indication of structural changes within a material. Hence the effect of gamma irradiation on light absorbance in CR-39 was also investigated.

EXPERIMENTAL PROCEDURE

The CR-39 was obtained from American Acrylics and Plastics, Inc. in the form of sheets 457×610×0.635 mm (18×24×0.025 in). Both surfaces of the sheets were protected by polyethylene film of thickness 60 μm. The sheets of CR-39 were laser-cut into pieces of dimensions 25.4 × 12.7 × 0.635 mm (1 × 0.5 × 0.025 in). It has been reported that laser cutting does not affect the track recording properties of CR-39.(4)

CR-39 detectors were exposed in pairs to doses of ^{60}Co gamma rays ranging from 0 to 280 kGy (0 to 28 Mrad). All gamma irradiations were performed using a ^{60}Co gamma cell.(5) Four unirradiated detectors were used to determine the background track density. A micrometer was used to measure the thickness of the CR-39 detectors before and after etching. The difference in thickness was used to determine the bulk etch rate. The bulk etch rate is defined as the thickness removed on one side per unit time. The accuracy of the micrometer was 2.54 μm (0.0001 in).

After gamma irradiation the CR-39 detectors were exposed to a known fluence of ^{252}Cf spontaneous fission neutrons (1.16×10^8 n/cm^2). The neutron irradiations were made by mounting the CR-39 detectors behind a circular polyethylene disc of thickness 0.77 mm which served as a proton irradiator. The polyethylene disc was attached to a rotating disc arrangement to ensure uniformity of incident neutron flux. The room temperature varied between 19°C and 23°C during all irradiations.

All detectors were etched at 70°C for 4 hours in 6.25N sodium hydroxide solution. The track counting was done using an optical microscope (American Optical Series 50) at 450 magnification. Only the tracks on the top surface of the detector were counted. For each detector twenty fields chosen at random were counted (total area of 9.68×10^{-3} cm^2). The average track density was determined for the two detectors. The average background track density was subtracted from the total track density to obtain the net track density.

The ultraviolet absorbance study was done with a calibrated Perkin Elmer double beam spectrophotometer, Coleman 124. The unetched detectors were placed one by one in the spectrophotometer with an unirradiated detector used as the blank. The absorbance at different wavelengths in the range from 190 to 370 nm was determined.

RESULTS AND DISCUSSION

Figure 1 shows the effect of pre-gamma irradiation on net track density in CR-39 irradiated with ^{252}Cf spontaneous fission neutrons. The net track density generally decreased with increasing gamma dose. The curve has been drawn only to aid the eye. The error bars represent the pooled standard error of the average track density for the two detectors.

Figure 2 illustrates the relationship between bulk etch rate and gamma dose. The curve has been drawn only to aid the eye. The error bars represent the accuracy to which the micrometer could be read. The bulk etch rate generally increases with increasing gamma dose. However, it must be pointed out that in the determination of the bulk etch rates, no correction was made for the swelling of the polymer during etching. A more reliable way of determining bulk etch rates is by measuring the diameters of tracks induced by fission fragments or heavy ions.(6,7) Other authors have also reported increasing bulk etch rates with increasing gamma doses.(1,2,3) Between 0 and 50 kGy (5 Mrad) the curve appears exponential. Beyond 60 kGy it appears linear. Zamani(3) has observed a similar behavior.

The irradiation of polymers with gamma rays results predominantly in ionization and excitation.(8) Subsequently the chemical bonds are ruptured yielding fragments of the large polymer molecules which may retain unpaired electrons from the broken bonds. The chemical and physical properties of the material may be altered due to the reaction of the free radicals. The polymer may either undergo scission, i.e., the polymer molecules may be broken into smaller fragments, or it may undergo cross linking, i.e., the molecules may be linked together to form larger molecules.

When polymers are irradiated with gamma rays the scission of macromolecules is shown macroscopically as a change in various properties of the polymer.(9) One such change is in the molecular weight. Scission results in a decrease in the molecular weight which in turn results in an increase in the bulk etch rate. By analogy an increase in the molecular weight results in a decrease in the bulk etch rate. This takes place if the molecules are joined together at points where the gamma radiation has created free radicals. In this case more than one kind of cross linking can take place. If cross linking takes place the molecular weight increases, resulting in a decrease in the bulk etch rate.

For CR-39 the increasing bulk etch rate with increasing gamma dose suggests the predominance of scission of the polymer when irradiated with gamma rays.

In chemical etching there is the simultaneous action of two processes, chemical dissolution along the particle trajectory at a linear rate V_t referred to as the track etch rate; and a general attack on the etched surface and on the interior of the track at a lower rate V_g referred to as the bulk or general etch rate.

The development of a particle track is the resulting effect of the two etch rates V_t and V_g.(3) The track etch rate V_t depends on the dose deposited along the particle trajectory. The bulk etch rate V_g depends inter alia on the background dose.

The etching efficiency for particles incident at a 2π solid angle (assumed to be isotropic) is given by the expression

$$\eta = 1 - \frac{V_g}{V_t}$$

where η, the etching efficiency is defined as the fraction of tracks intersecting a given surface that are etched on the surface under specified etching conditions.(10)

The etching efficiency deals primarily with geometrical requirements for revealing the tracks that are present. Changes in the ratio V_g/V_t affect the etching efficiency and in turn the track density. If the ratio V_g/V_t increases, η decreases and the track density decreases. If the background dose is the same as the dose deposited along the particle trajectory i.e., $V_g = V_t$, then the etching efficiency reduces to 0 and no track is revealed. This could be the explanation for the decreasing track density from 0 to 30 kGy as is seen in Figure 1. The bulk etch rate also increases in this dose range as is evident from Figure 2. This would result in a decreasing etching efficiency, and hence a decreasing track density with gamma dose.

Figure 3(a) shows the tracks in CR-39 exposed to a known neutron fluence (1.16×10^8 n/cm^2). Figures 3(b), 3(c), 3(d), 3(e) and 3(f) show the tracks in CR-39 exposed to the same neutron fluence but pre-irradiated with gamma doses of 3, 25, 67, 140 and 280 kGy (0.3, 2.5, 6.7, 14 and 28 Mrad).

The photomicrographs in Figure 3 show the apparent decrease in track density and the increase in background effects with increasing gamma doses in the range of 0 to 25 kGy (2.5 Mrad). For gamma doses greater than 30 kGy (3 Mrad) [Figures 3(d), 3(e) and 3(f)], the photomicrographs show the increasing severity of the background effects and the loss of well-defined tracks. This is why tracks could not be counted for gamma doses greater than 30 kGy. The track diameters also seemed to increase with gamma dose as is indicated by the photomicrographs. The mechanism of track etching demands that track diameters bear a direct proportionality to the bulk etch rate. The bulk etch rate did increase with gamma dose, as can be seen from Figure 2.

The absorption of ultraviolet light by unetched gamma irradiated CR-39 was investigated in the wavelength range of 190 to 370 nm. The results are shown in

Figures 4, 5 and 6. The absorbance is defined as log I_0/I where I_0 is the intensity of the transmitted light through an unirradiated sample taken as reference and I is the intensity of the transmitted light through the irradiated sample.

The ultraviolet absorption spectra for CR-39 samples irradiated with various gamma doses is shown in Figure 4. Ultraviolet absorption in a material is due to electron transitions that can take place in the material. The absorbing species may be initially present in the material or could be formed by degradation of the material.(3) In solid substrates electron traps can also contribute to absorption. Generally the interaction of gamma rays with a polymer results in the breaking of bonds and displacement of electrons and atoms.(11) These displaced electrons and ions migrate through the material until they are trapped somewhere in the lattice, thus leaving deficient regions. The electronic and ionic configurations thus formed may cause preferential light absorption at particular wavelengths.

The interaction of gamma radiation with polymers could also produce organic species, which would also play a significant role in the absorption of ultraviolet light. The peaks in Figure 4 are quite broad indicating the likely formation of more than one type of organic species or centers. As the relative composition of the components of the organic species or centers changes the peak broadens and shifts in wavelength.

In Figure 4 we also see that there is a deviation from zero absorbance when unirradiated CR-39 samples are used both in the reference beam and in the sample beam. This deviation is not very significant and can be caused by minor variations in the thicknesses of the samples (which we observed), and from the fact that the samples were not always perpendicular to the incident UV beam. Hence the absorbance studies are not quite quantitative. A small correction needs to be made for these variations.

The total area under the curve of absorbance versus wavelength in the range of 220 to 370 nm (Figure 4) is related to the number of centers or absorbing species contributing to the absorption band.(11,12) This in turn is related to the absorbed dose in the CR-39 sample. The areas under the curves in Figure 4 were determined with a planimeter and are shown in Figure 5 plotted against the corresponding gamma doses. The area-gamma dose relationship appears to be quadratic from 0 to 40 kGy and linear for higher gamma doses.

Figure 6 shows the absorbance of UV light at specific wavelengths by CR-39 for various gamma doses. Here again it can be observed that beyond 40 kGy the absorbance increases linearly with gamma dose, but the increase is less pronounced from that between 0 and 30 kGy. Sayed and co-workers have also reported the effect of gamma radiation on ultraviolet light absorbance in CR-39.(13)

From Figures 4, 5 and 6 the changes in light absorbance clearly indicate structural changes in CR-39 when irradiated with gamma radiation.

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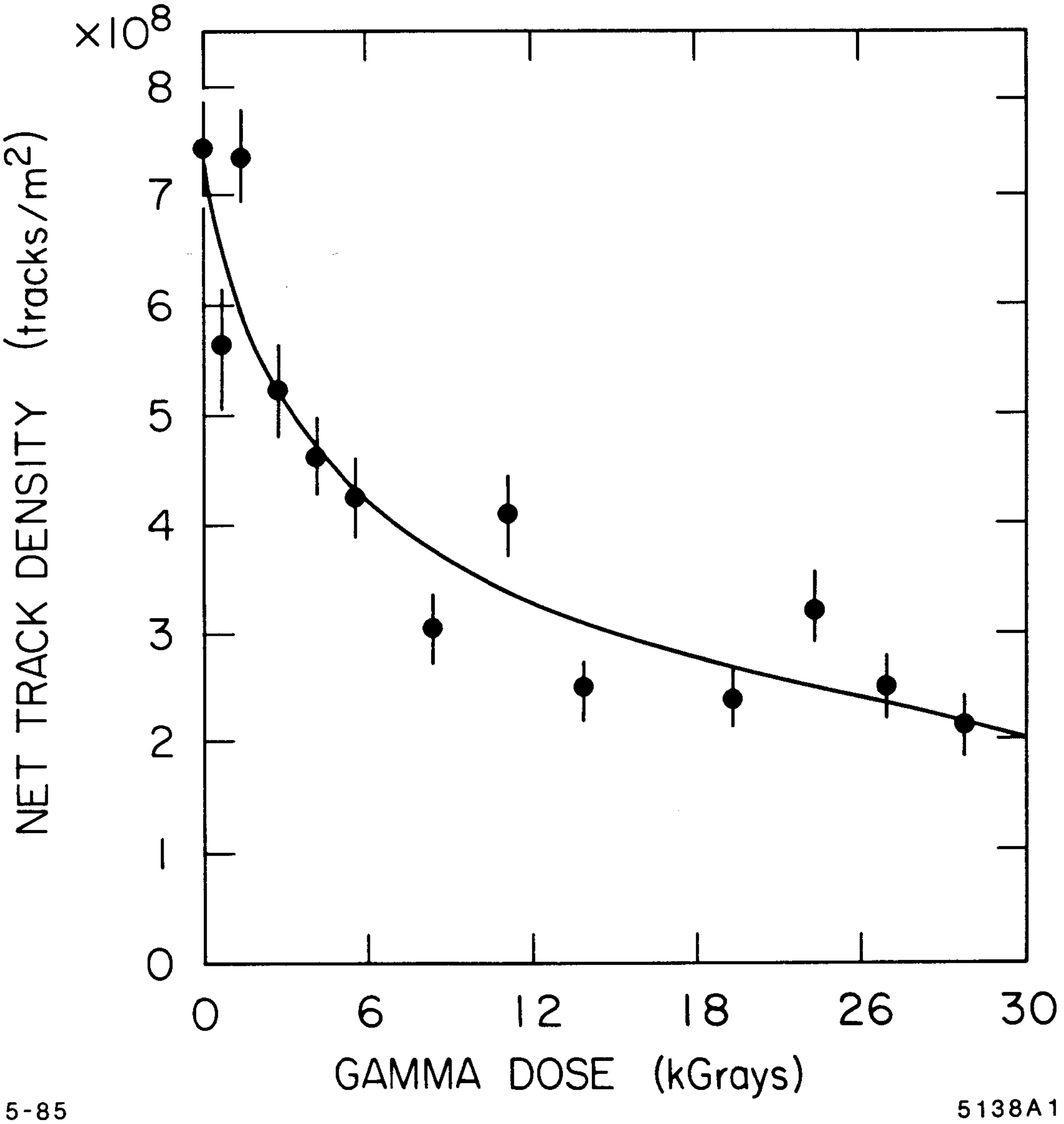
FIGURE CAPTIONS

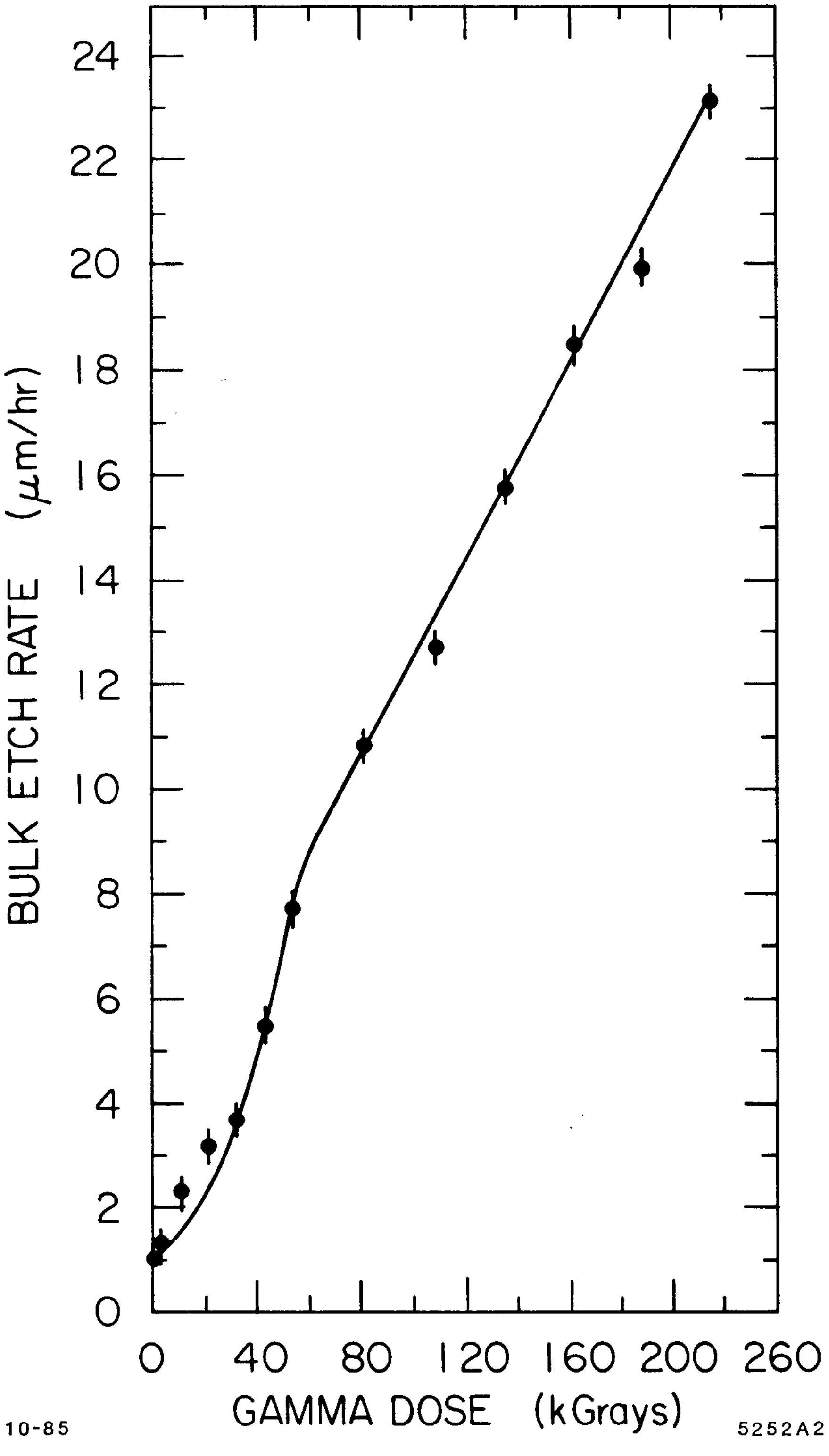
1. Effect of pre-gamma irradiation on track density in neutron irradiated CR-39.
2. Effect of gamma radiation on bulk etch rate in CR-39.
3. Photomicrographs showing effect of pre-gamma irradiation with ^{60}Co on track density in neutron irradiated CR-39.
4. Absorbance of UV light by CR-39 for various gamma doses.
5. Variation of area of absorption band as a function of gamma dose.
6. Absorbance of UV light at specific wavelengths by CR-39 for various gamma doses.



5-85

5138A1

Fig. 1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6
8
10
12
14
16
18
20
22
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120
160
200
260
10-85
5252A2

Fig. 2

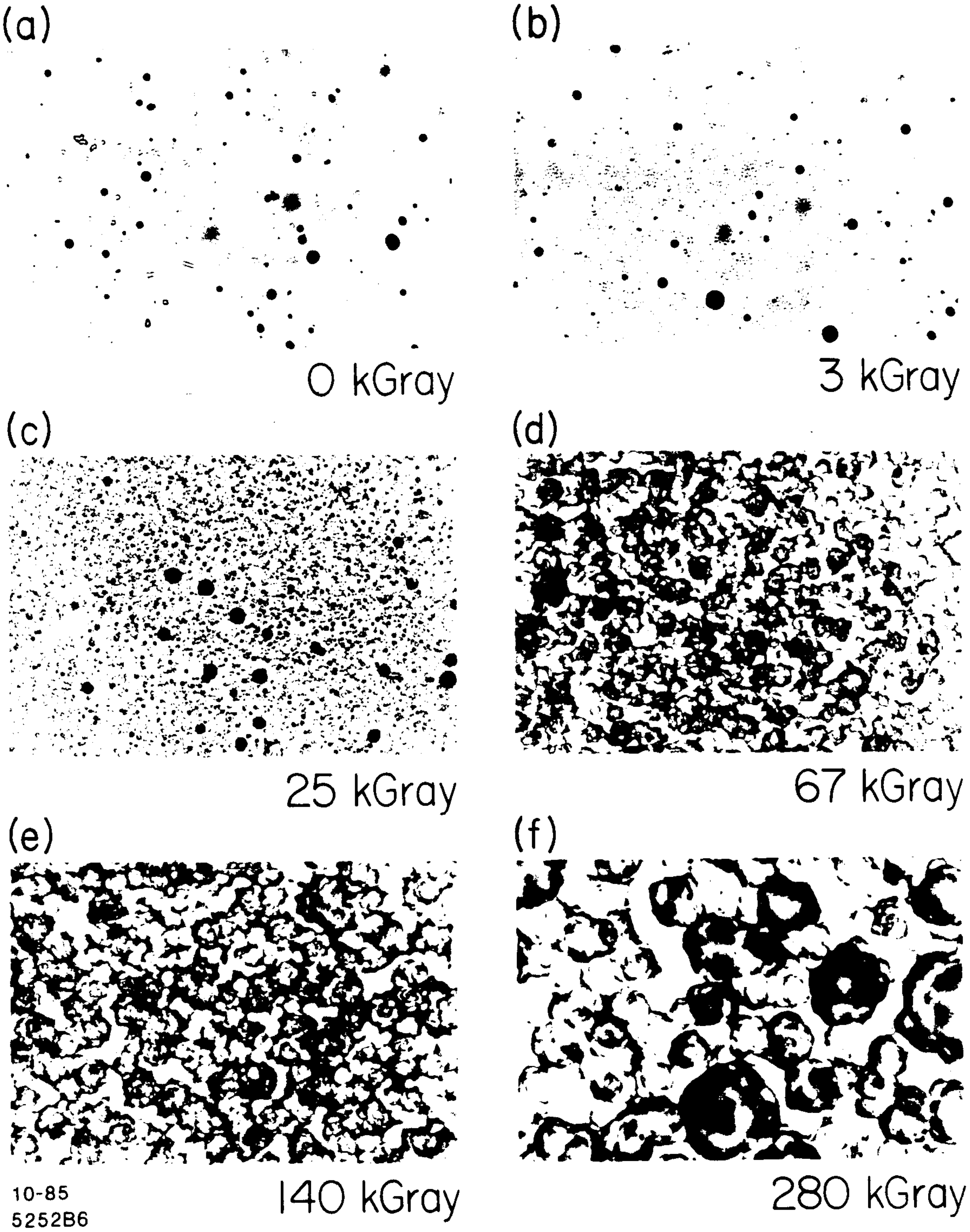

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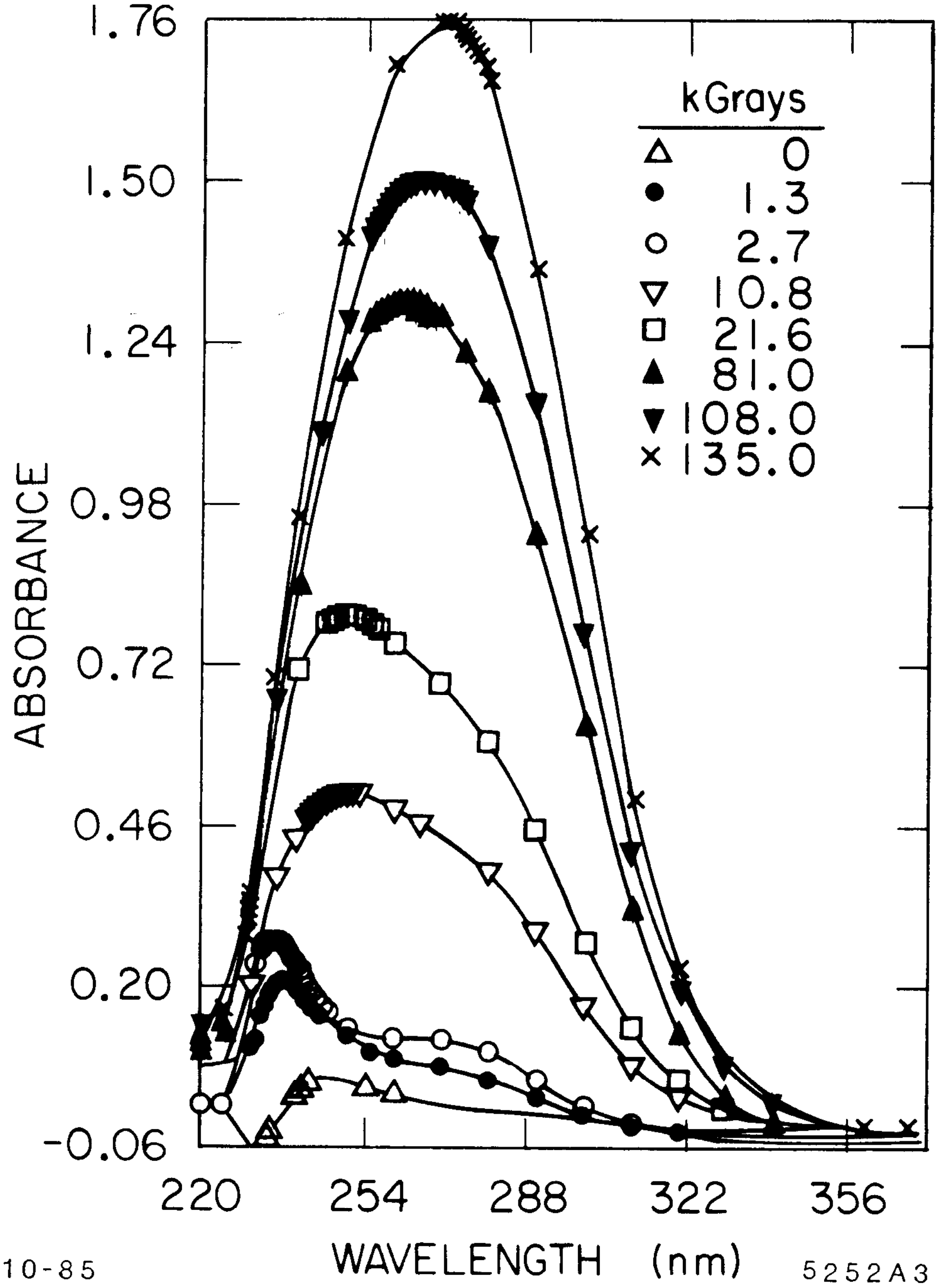
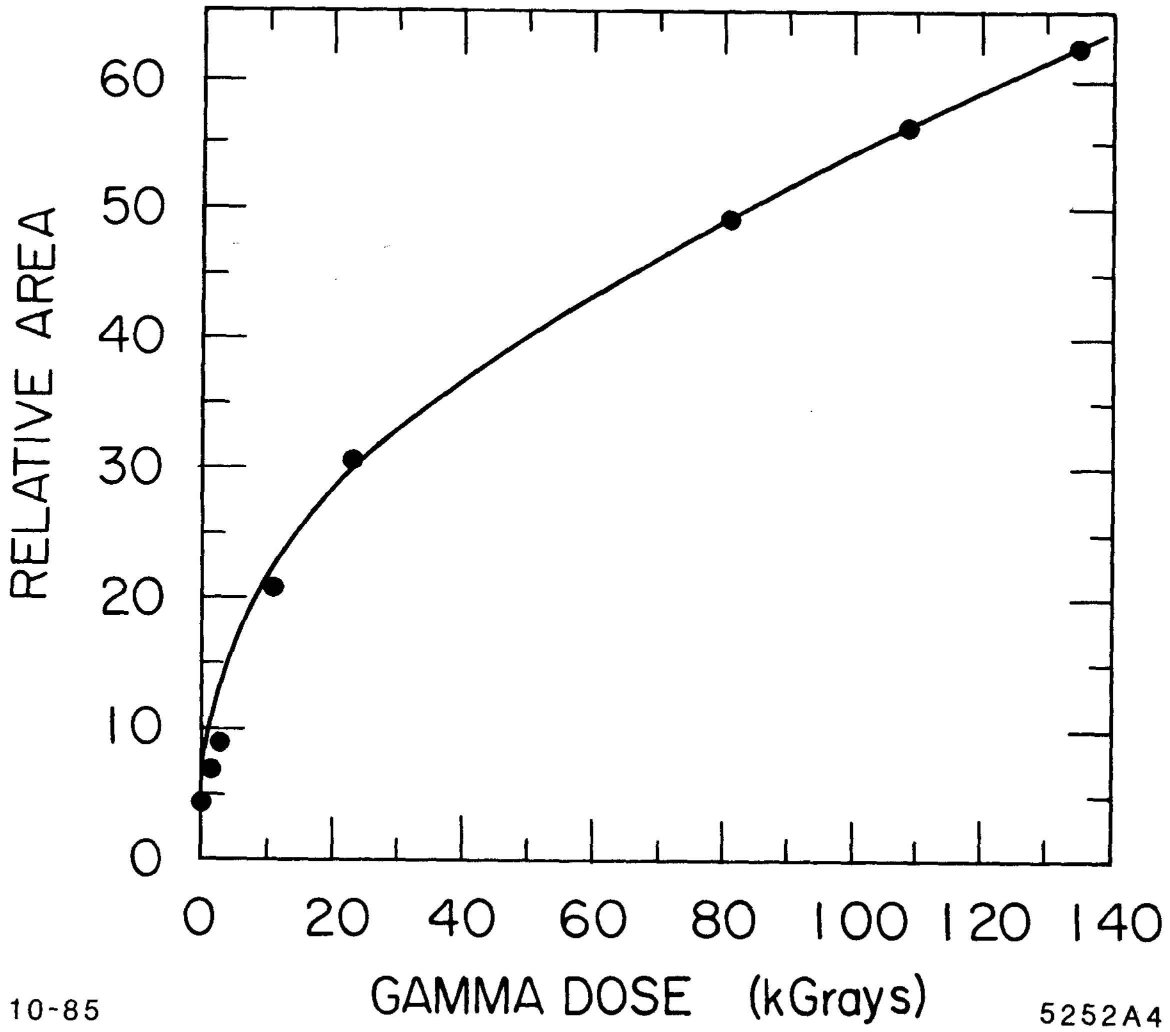
Fig. 3



10-85 5252A3

Fig. 4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50
40
30
20
10
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100
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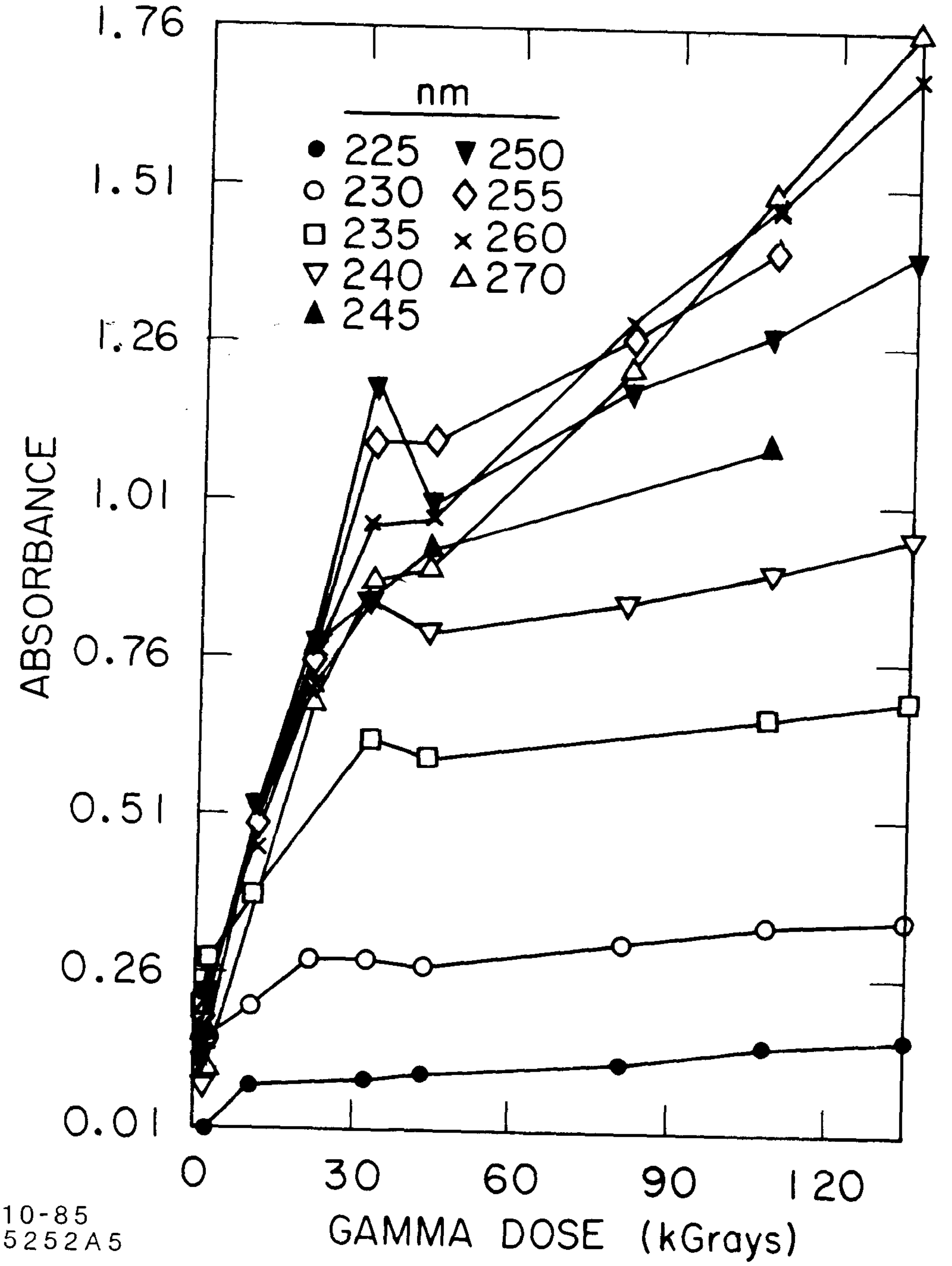
140
GAMMA DOSE (kGrays)
10-85
5252A4

Fig. 5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0.76
0.51
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0.01
ABSORBANCE
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90
120
GAMMA DOSE (kGrays)
nm
● 225
○ 230
□ 235
▽ 240
▲ 245
▼ 250
◇ 255
× 260
△ 270
10-85
5252A5

Fig. 6