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EFFECT OF ANNEALING ON TRACK DENSITY IN UNIRRADIATED AND GAMMA IRRADIATED CR–39 WHEN USED FOR FAST NEUTRON DETECTION*

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ABSTRACT

The effect of annealing on track density in unirradiated CR–39 and gamma irradiated CR–39 neutron detectors was investigated.

One group of CR–39 detectors (25.4 × 12.7 × 0.635 mm) was irradiated with 1.3 kGy of ^{60}Co gamma rays. The other group of detectors was not irradiated with gamma radiation. The two sets of detectors were then annealed at temperatures ranging from room temperature (20°C) to 150°C for 30 minutes. The detectors were then overlaid with 0.77 mm of polyethylene and exposed to ^{252}Cf spontaneous fission neutrons. Eight detectors were used as controls for the determination of background track density.

All detectors were then etched in 6.25 N sodium hydroxide solution at 70°C for 4 hours. The track densities were determined with an optical microscope.

In the case of gamma irradiated detectors the net track density increased non-linearly with temperature until 150°C. For the unirradiated detectors the net-track density initially decreased with increasing temperature up to 60°C, increased between 60°C and 120°C then sharply decreased between 120°C and 130°C, remaining constant thereafter.

Ultraviolet spectrophotometry measurements showed significant changes in light absorbance indicating heat induced structural changes in the detector for both annealed CR–39 detectors and gamma irradiated annealed CR–39 detectors.

INTRODUCTION

CR–39 is a polymeric nuclear track detector which is being increasingly used for fast neutron dosimetry. An ideal dosimeter should retain most of the dose information under various environmental conditions. If the environmental effects on the dosimeter are unknown then the estimate of the dose could lead to erroneous results. During use track detectors can be exposed to temperatures higher than ambient levels. Elevated temperatures can induce changes in the bulk detector material and in the latent damage tracks. The effect of temperature on the material and on track registration depends on whether the material has been subjected to heating prior to or after the formation of radiation induced latent damage tracks. Thus there are two temperature effects, the so called pre- and post-irradiation annealing effects (Ogura and Tamai (1982)). Pre-irradiation annealing alters the property of the bulk material and post-irradiation annealing affects both the bulk material as well as the latent damage tracks.

Some aspects of the effect of temperature on the registration properties of CR–39 have been reported (Al Najjar *et al.* (1982); Gruhn *et al.* (1980); O'Sullivan *et al.* (1981); Jamil *et al.* (1981); O'Sullivan *et al.* (1984); Hamasaki *et al.* (1984); and Wong and Field (1984)).

Ultraviolet absorbance studies are powerful techniques for the indication of structural changes within a material. Hence the effect of annealing on ultraviolet light absorbance in unirradiated and gamma irradiated CR–39 was also investigated.

EXPERIMENTAL PROCEDURE

The CR–39 was obtained from American Acrylics and Plastics, Inc. in the form of sheets $457 \times 610 \times 0.635$ mm. 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 $254 \times 12.7 \times 0.635$ mm. It has been reported that laser cutting does not affect the track recording properties of CR–39 (Kukreja *et al.* (1984)).

One group of CR–39 detectors was irradiated with 1.3 kGy of ^{60}Co gamma rays. The gamma irradiation was performed using a ^{60}Co gamma cell (Ipe (1984)) at an exposure rate of 46 Grays/minute. The other group of detectors was not irradiated with gamma radiation. After an interval of at least one day pairs of detectors from each of the two sets were annealed at temperatures ranging from room temperature (20°) to 150° for 30 minutes. The next day the 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 detector behind a circular polyethylene disc of thickness 0.77 mm, which served as a proton radiator. The polyethylene disc was attached to a rotating disc arrangement to ensure uniformity of incident neutron flux. Four unannealed unirradiated (by gamma or neutrons) detectors were used as controls for each group for the determination of background track density.

All detectors were etched at 70°C for 4 hours in 6.25 N sodium hydroxide solution. All track counting was done using an optical microscope (American Optical Series 50) at 450 magnification. Usually the track counting was done one day after the etching. Only the tracks on the top surface of the detector were counted. For each detector twenty fields chosen at random were counted. The average track density was determined for each pair of detectors. The average background track density was subtracted from the total track density to obtain the net track density.

The ultraviolet light absorbance study was done with a calibrated Perkin Elmer double beam spectrophotometer Coleman 124. In this case the two groups of detectors were annealed under the same conditions as before except that they were not exposed to neutrons. 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

Figures 1 and 2 show the effect of annealing temperature on net track density in unirradiated and gamma irradiated CR–39 respectively. The curves have been drawn only to aid the eye. The error bars represent the pooled standard error of the average track density for the two detectors. The neutron sensitivity for unirradiated CR–39 at 20°C is 6.6×10^{-4} tracks/neutron for ^{252}Cf spontaneous fission neutrons. This compares favorably with neutron sensitivities reported by other investigators provided the differences in etching conditions, radiator thicknesses and neutron sources are taken into account (Gomaa and Kasim (1980), Benton *et al.* (1981)).

In Figure 1 the net track density exhibits an unusual pattern of behavior. Between 20°C and 60°C there is a 30% decrease in track density. From 60°C to 120°C there is over a 100% increase in track density. From 120°C to 130°C there is a sharp decrease of 65% in track density. Beyond 130°C the track density remains the same up to 150°C. Materials heated beyond 150°C for 30 minutes showed evidence of heat distortion and discoloration.

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 etching efficiency for particles incident at a 2π solid angle 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 under specified etching conditions (Fleischer *et al.* (1975)). 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.

Higher annealing temperatures often result in the degradation of the polymer. The material is then considerably softened resulting in an increase in the bulk etch rate which would result in a decrease in the etching efficiency. This could be a possible explanation for the decrease in track density between 120° and 130°. An increase in the bulk etch rate was also observed in this temperature range (Ipe (1984)). 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 annealing and etching. Other investigators have also observed an increase in the bulk etch rate in this temperature range (Ogura and Tamai (1982); Jamil *et al.* (1981)).

The nature of the mechanisms that cause changes in track density in the other temperature ranges are not fully understood. Pre-irradiation annealing alters the internal morphology of the polymer (Aframian (1978)). The polymer chain spacings are probably rearranged thus changing the detection thresholds. This may account for the change in track density in the other temperature ranges.

In Figure 2 the net track density for gamma irradiated CR–39 appears to increase between 20°C and 141°C. However the net track density at 20°C for the gamma irradiated CR–39 is 30% less than that for unirradiated CR–39. At 120°C the net track density for gamma irradiated CR–39 approaches that of the unirradiated CR–39 at 20°C. At 141°C the track density for gamma irradiated CR–39 is about 30% greater than that for unirradiated CR–39 at 20°C. It is quite evident that there is a significant interaction effect (between gamma dose and temperature) which causes the CR–39 to respond quite differently than when it is subjected to temperature alone. Studies on the bulk etch rate indicated that it increased non-linearly (much like the curve in Figure 2) with temperature between 20°C and 170°C for gamma irradiated CR–39 (Ipe (1984)).

The absorption of ultraviolet light by unetched CR–39 was investigated in the range of 190 to 370 nm for both unirradiated and gamma irradiated CR–39. The results are shown in Figures 3 and 4. The absorbance here is defined as log I_0/I where I_0 is the intensity of the transmitted light through an unannealed sample and I is the intensity of the transmitted light through the annealed sample.

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 the degradation of the material. In solid substrates electron traps can also contribute to absorption.

The absorbance was found to peak at about 240 nm for both unirradiated and gamma irradiated CR–39 at various annealing temperatures. The increase of absorbance with increasing temperatures may indicate the increased production of absorbing species. In both figures the absorbance increases with temperature beyond 112°C. Between 20°C and 112°C the absorbance goes through maxima and minima. In both cases it appears that some kind of transition takes place at 80°C and 112°C which results in the loss of absorbing species from the material, perhaps in a volatile form. Another possible explanation is that the main absorbing species is some combination of temporary and permanent species with short and long lifetimes respectively.

The absorbance at 98°C is higher than that at 60°C for gamma irradiated CR–39 unlike that of unirradiated CR–39. Figures 3 and 4 indicate that some structural compositional changes are taking place at different temperatures and this could be the cause for the changing track density with temperature for both unirradiated and gamma irradiated CR–39.

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FIGURE CAPTIONS

Figure 1: Effect of annealing temperature on track density in unirradiated CR–39.

Figure 2: Effect of annealing temperature on track density in gamma irradiated CR–39.

Figure 3: Absorbance of UV light at 240 nm by unirradiated CR–39 for various annealing temperatures.

Figure 4: Absorbance of UV light at 240 nm by gamma irradiated CR–39 for various annealing temperatures.

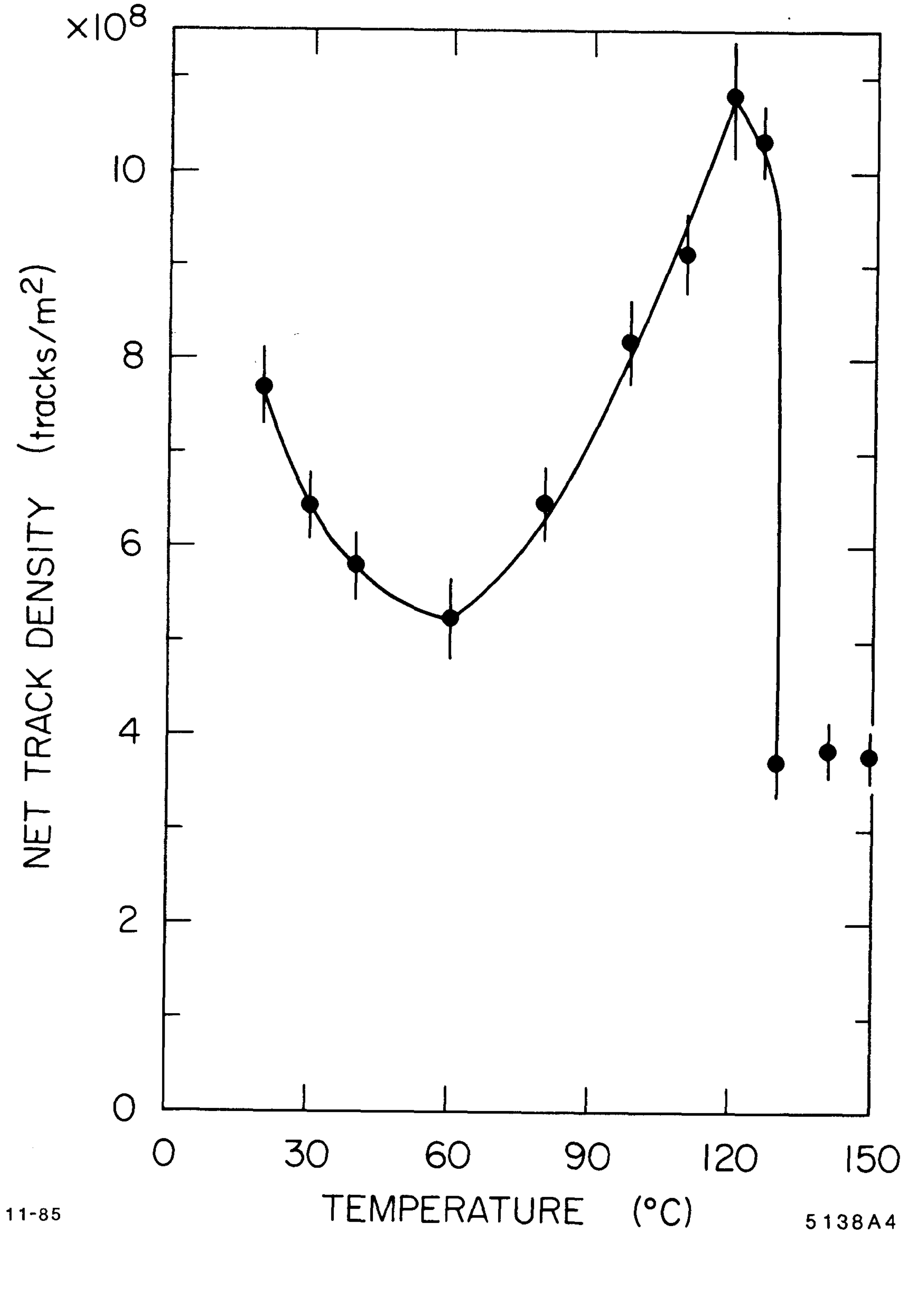

×10^8
10
8
6
4
2
0
NET TRACK DENSITY (tracks/m^2)
0
30
60
90
120
150
TEMPERATURE (°C)
11-85
5138A4

Fig. 1

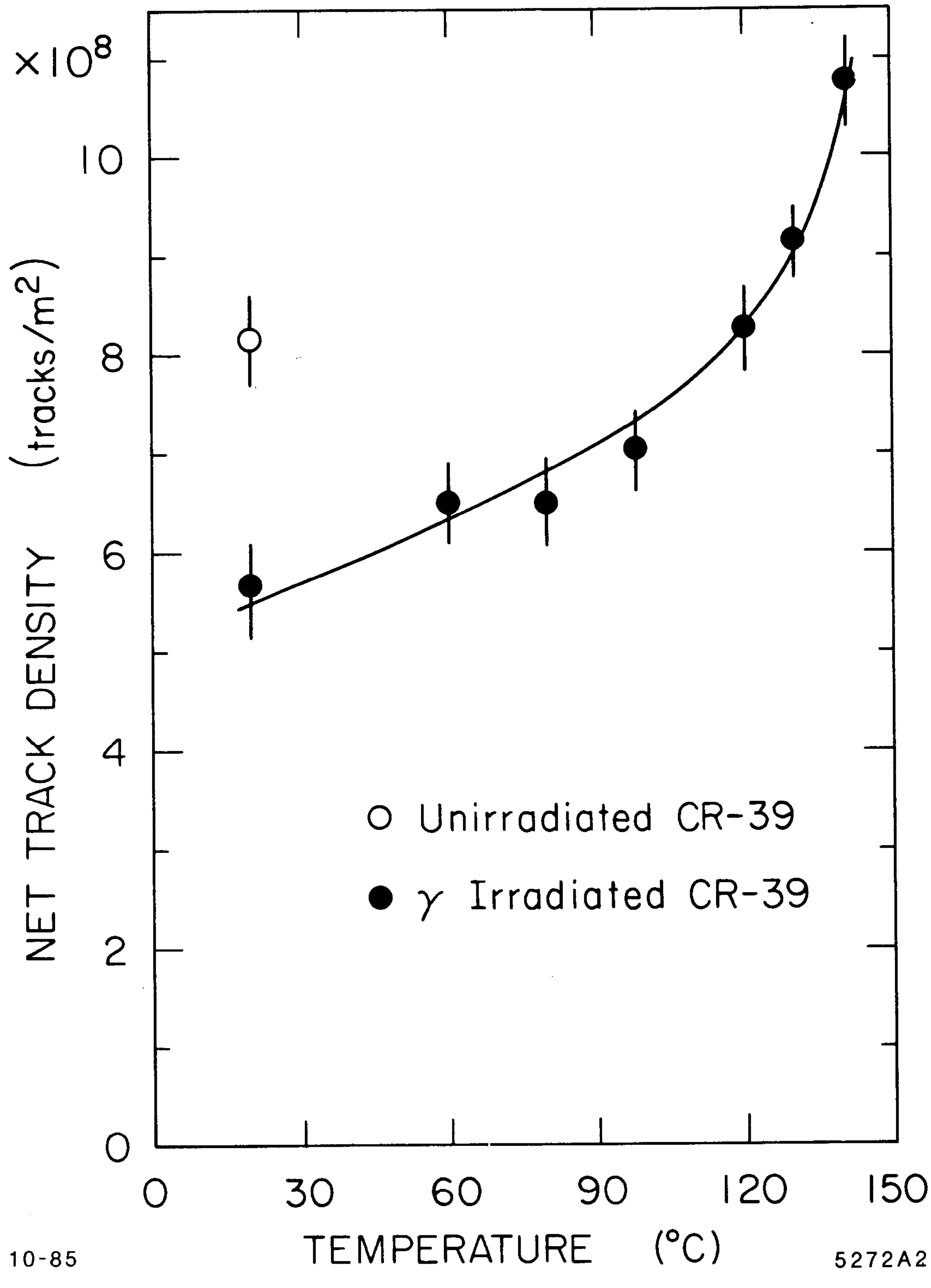
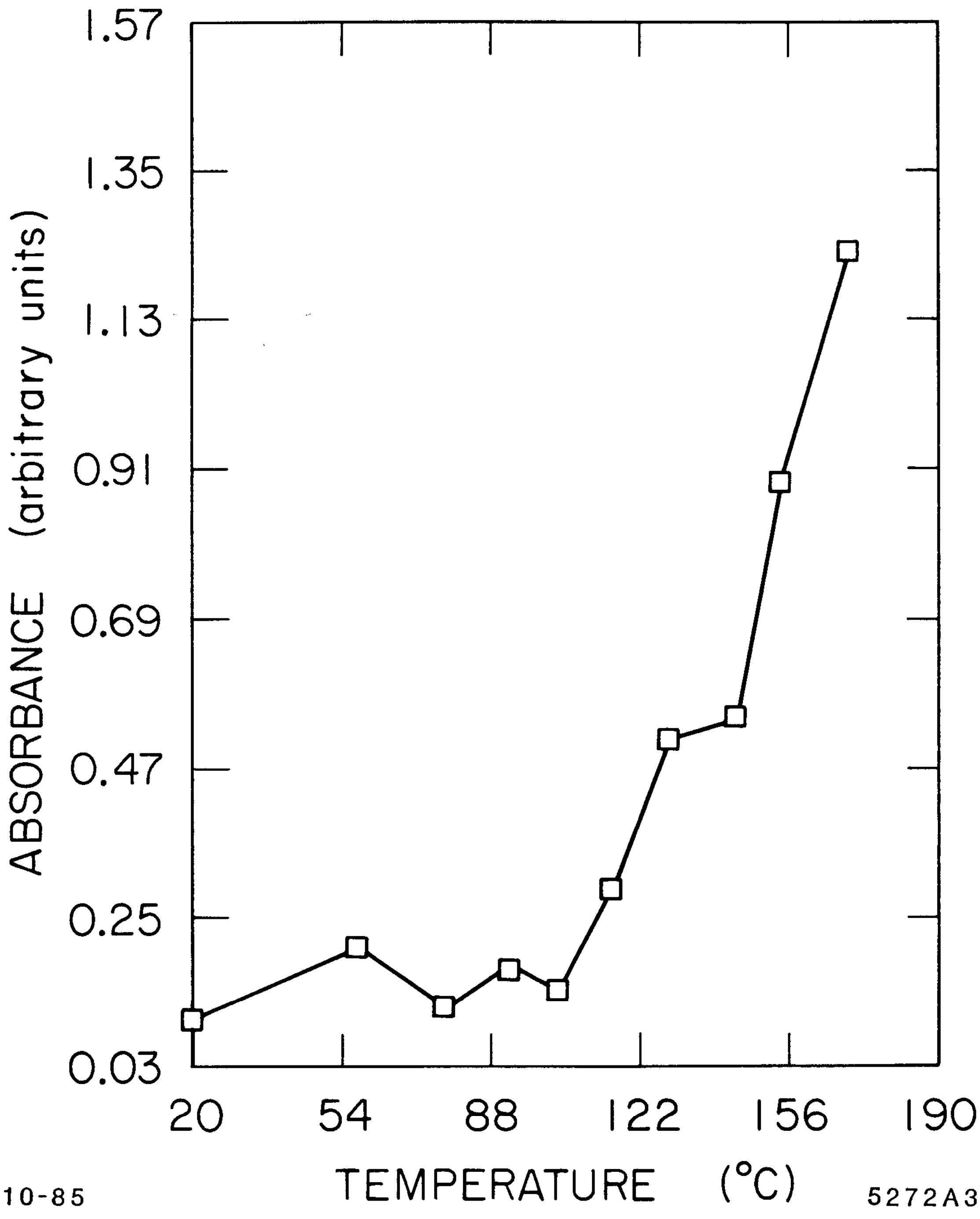

×10^8
NET TRACK DENSITY (tracks/m²)
0
2
4
6
8
10
0
30
60
90
120
150
TEMPERATURE (°C)
○ Unirradiated CR-39
● γ Irradiated CR-39
10-85
5272A2

Fig. 2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1.35
1.13
0.91
0.69
0.47
0.25
0.03
ABSORBANCE (arbitrary units)
20
54
88
122
156
190
TEMPERATURE (°C)
10-85
5272A3

Fig. 3

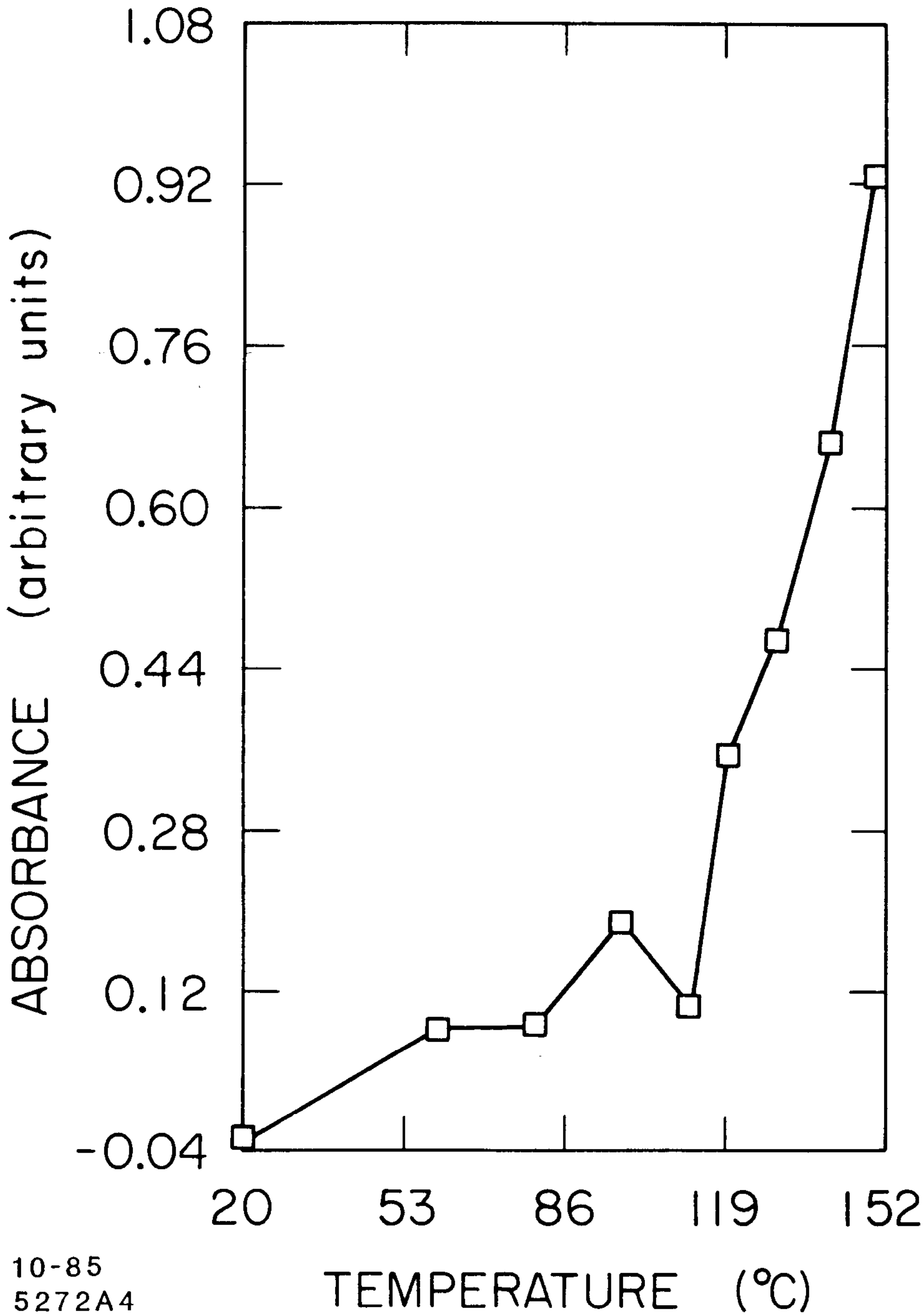
1.08
0.92
0.76
0.60
0.44
0.28
0.12
-0.04
ABSORBANCE (arbitrary units)
20
53
86
119
152
TEMPERATURE (°C)
10-85
5272A4

Fig. 4