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Study of the Latent Fading of NTA Film and Track Etching Detectors at Various Temperatures and Humidities

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STUDY OF THE LATENT FADING OF NTA FILM AND TRACK ETCHING DETECTORS AT VARIOUS TEMPERATURES AND HUMIDITIES

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Abstract

After exposure to ^{252}Cf fission neutrons the latent fading of nuclear track detectors was investigated for storage periods up to 30 days at temperatures of 5°C, 25°C, and 50°C and relative air humidities of 0 %, 53 % and 97 %. Fading results are discussed for

- Kodak NTA films in the original wrapping and in an aluminium-plastic sealing,
- cellulose acetate and Makrofol E for the detection of neutron induced recoils and α-particles,
- Makrofol E and KG in contact with a thorium foil for the detection of fission fragments.

NTA film and cellulose acetate show a high fading up to more than 50 % after a period of 3 weeks, but no tracks at higher temperature and humidity. Under the extreme conditions the fading of Makrofol was found to be 23 % after a 3 weeks period, but no fading was found under laboratory conditions for a period of 2.5 years.

Zusammenfassung

Nach einer Bestrahlung mit ^{252}Cf-Spaltneutronen wurde das Latentbildfading von Kernspurdetektoren bis zu Lagerungszeiten von 30 Tagen bei Temperaturen von 5°C, 25°C und 50°C und bei einer relativen Luftfeuchte von 0 %, 53 % und 97 % untersucht. Es werden Fadingergebnisse diskutiert für

- Kodak NTA Filme in Originalverpackung und in einer Aluminium-Plastik-Verschweißung,
- Zelluloseazetat und Makrofol E für den Nachweis von neutroneninduzierten Neutronen und α-Teilchen,
- Makrofol E und KG in Kontakt mit einer Thoriumfolie für den Nachweis von Spaltfragmenten.

NTA Film und Zelluloseazetat zeigen ein hohes Fading bis zu mehr als 50 % nach 3 Wochen, aber keine Spuren mehr bei hoher Temperatur und Luftfeuchtigkeit. Unter extremen Verhältnissen betrug das Fading von Makrofol 23 % nach 3 Wochen, unter Laborbedingungen wurde jedoch nach 2,5 Jahren kein Fading gefunden.

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Fig. 36 Relative number of track vs. storage time for Kodak NTA, Makrofol and cellulose acetate, stored at 50°C and 97 % R.H.

1. Introduction

The registration of nuclear tracks in photographic emulsions has been used over a period of 20 years as the only method of measuring fast neutrons in personnel dosimetry. Personnel neutron monitoring based on recoil proton registration became the major application of nuclear track emulsion in the field of health physics. The nuclear track emulsion, which has been widely used for personnel neutron monitoring, is the special fine-grained NTA Kodak Personnel Neutron Monitoring Film, Type A. Several investigations have shown that the application of the Kodak NTA film is limited by the fact that the response to fast neutrons depends on the neutron energy, direction of radiation incidence, track counting, to a high degree on latent fading, particularly at higher temperatures and humidities, and on the fogging of the emulsion by gamma irradiation. Therefore, the application of track etching methods in solid state detectors becomes the most widespread tool in fast neutron dosimetry as an alternative to the nuclear track emulsion as well as to other measuring techniques using threshold detectors. One of the most important properties of solid state track etching detectors is the insensitivity to gamma radiation and the latent track stability. Following the first studies of Fleischer, Price and Walker [10] in the early sixties track etching detectors have now been used as a standard technique to measure neutron fluence in the field of neutron sources, where reproducibility and accuracy of track counting are one of the important detector characteristics.

The latent fading of recoil protons in the NTA emulsion as well as of damage regions along heavy charged particle tracks in solid-state detectors is responsible for a more or less significant lower or higher loss of information, particularly on long-term monitoring under different environmental conditions. One of the reasons for latent track instability is the influence

of temperature, air humidity and oxygen. In the period between exposure and development, the latent tracks can undergo several changes by storage under various conditions of temperature and relative humidity. From the practical point of view, the influence of environmental conditions on the latent fading affects the significance and accuracy of neutron fluence measurements, calibrations in neutron fields, spectra measurements by threshold detectors as well as of routine and accidental personnel monitoring.

During the past 10 years many investigators have studied the fading properties of the different track detectors which were used until that time in their laboratories. Up to now, one can find a lot of information about fading but there are no complete results of studies showing the fading kinetics with the same parameters of environmental conditions for all detectors of interest. In a recent report Becker [20] gives a literature survey of measured results found for the NTA film and other dosimeters by different authors within the past 10 years. However, the significance of such investigations depends on the requisites of the laboratory and the practical conditions of development and track counting as given in Tab. 1 for the NTA film by Cavallini and Busuoli [11]. It appears from this table that, for the same type Kodak NTA, the results given by 18 different Laboratories are varying between "no fading" and 90 % of fading within 30 days. Jasiak and Musialowicz [12] found, according to some data obtained by several investigators, that the number of recoil proton tracks decreases by 5 to 100 %, depending on various wrappings of the NTA film and storage conditions between irradiation and development as shown in Tab. 2. To interpret these results, we must consider different environmental conditions as well as different materials and techniques used for film packing. For these reasons, a comparison of the results is of minor value. On the other hand fading results found at high temperatures provide no practical information about the fading influence under routine environmental conditions.

Table 1: Results of fading experiments in Kodak NTA after Cavallini and Busuoli [11]

Laboratory	Influence of Fading
A	no, within the measurement errors
B	no, within the measurement errors
C	no, in particular packing
D	no, for the packing
E	no, in 30 days for the packing
F	5 % in 60 days
G	10 % after 2 months
H	6-7 % in 4 weeks in special wrapping
I	7-8 % after 4 weeks in special wrapping
L	16 % in 3 weeks
M	20 % in 30 days
N	20 % in 30 days
O	30 % in 30 days (wrapped) 90 % in 30 days (unwrapped)
P	20 % in 15 days
Q	20 % in 15 days
R	50 % in 30 days
S	50 % in 15 days
T	50 % in 15 days

Table 2: Results of track fading investigations after Jasiak and Musialowicz [12]

Author	Kind of wrapping	Period of storage of exposed films	Environmental conditions of exposed film storage	Losses in number of tracks in %
1	2	3	4	5
Cheka [1]	original factory wrapping only	64 days 16 days 5 days	in desiccator 50% relative humidity at 24°C unmodified atmosphere	12 12 12
Cusimano [2]	sealed in mylar original factory wrapp. only	42 days 42 days	average relative humidity 30%, 22°C	no significant regression minimal drop including in the measuring ranges
Hagsgard and Widell [3]	original factory wrapping only	14 days 60 days	50% relative humidity temp. 20°C	20 50
Carthy and Mejdhal [4]	original factory wrapping only	3 weeks 8 weeks 3 weeks 8 weeks	in desiccator at temp. 16÷25°C under normal room conditions	16 22 16 76
Dresel [5]	no additional wrapping	2 days 2 days	in refrigerator temp. +2°C÷4°C temp. 55°C 75% relative humidity	0 100
Portal [6]	aluminium-polyethylene foil prior desiccation of films and wrappings without additional wrapping	3 months 1 week	85% relative humidity temp. 25°C 85% relative humidity temp. 25°C	15 75
Amadesi *et al.* [7]	without additional wrapping	60 days 60 days	relative humidity 5% temp. 20°C relative humidity 70%	no significant drop 100
Turner [8]	no additional wrapping	3 weeks 8 weeks	50% relative humidity	approx 20 55
Kahle *et al.* [9]	no additional wrapping	7 days 14 days	50±5% relative humidity temp. 24,5°±0,5°C	33*) 17**) 56*) 30**) neutrons *) of 0.9 MeV **) of 1.3 MeV
Zelac [30]	no additional wrapping		typical use conditions	average 2.6 per day
Jasiak and Musialowicz [12]	original factory wrapping only; additional polyethylene wrapping; additional polyethylene + prior desiccation of films and holders	30 days	60% relative humidity temp. 22°C	45 30 25

Using non-photographic track detectors, we have to consider the kind of plastic foils, the detector combination, the etching conditions, and the kind of counting technique, i.e. microscope or spark counter. All these parameters may affect the amount of latent track fading which we get experimentally. Published fading results are, therefore, only valid for the laboratory techniques applied with a special detector combination. Contrary to the NTA film, these detectors are no standard detectors which we can obtain from the manufacturer. But even the Kodak NTA film is home packed in every laboratory under "similar conditions" and there are no standard techniques for developing nuclear track films.

From the practical point of view it was, therefore, of interest to carry out fading experiments with the different types of track detectors presently used with the conditions of temperatures and relative humidities remaining the same. The aim of this study is to investigate the effects of these environmental conditions on the number of tracks and hence to compare the detector properties. The results of these investigations should be of general use, to find out the special fading characteristics of a track detector in the temperature range 0-50°C and for relative humidities of 0 % - 97 %. From these experiments also an intercomparison is possible of the photographic track detector and fission fragment track detectors as well as of the detection of neutrons using neutron induced recoils in plastic foils.

2. Description of requirements of the fading experiments

2.1 Detectors

The different types of detectors studied were Kodak Personnel Neutron Monitoring Film Type A (Kodak NTA) in the original wrapping as well as welded in aluminium-plastic foil, detector combinations of Makrofol KG (thickness 12 µm) with thorium, Makrofol E (thickness 300 µm) with thorium, as well as foils

of Makrofol E and cellulose acetate without targets. With these detectors the following types of particles were recorded:

- Recoil protons in the Kodak NTA film for the detection of fast neutrons > 0.7 MeV
- Fission fragments in Makrofol KG with thorium and Makrofol E with thorium respectively, for the detection of fast neutrons above a threshold of 1.2 MeV
- Neutron induced recoils and α-particles in Makrofol E
- Neutron induced recoils and α-particles in cellulose acetate.

The energy dependence of the detectors used in the experiment is given in Figs.1-3. The energy dependence of the Kodak NTA film was calculated [13] based on the hydrogen content of the film materials for recoil protons with energies Ep > 0.425 MeV (lowest track length of 4 silver grains) and found experimentally by Lehmann [14] and Piesch [15]. For the detection of neutron induced recoils in the polycarbonate foil Makrofol E experimental results by Józefowicz [16] are presented in Fig. 2, which were found for monoenergetic neutrons after a prolonged etching period of 7 hours in 6.25 N KOH (60°C). In Fig. 3 the energy dependence of the neutron cross section is shown for threshold reactions in the energy range of fast neutrons. In the experiment a thorium foil was used which shows a saturation thickness for fission fragments.

The dose range is 20 mrem - 10 rem for the counting of recoil protons in the NTA film, 5 - 5000 rem for the microscopical counting of neutron induced recoils in Makrofol E [17], and 100 mrem - 5000 rem for fission fragment etch pits, using a spark counter for low track density.

2.2 Environmental conditions

For the fading experiments three different temperatures were chosen: 5°C, 25°C, and 50°C. The temperature was maintained constant by using a thermoregulated chamber. The relative air

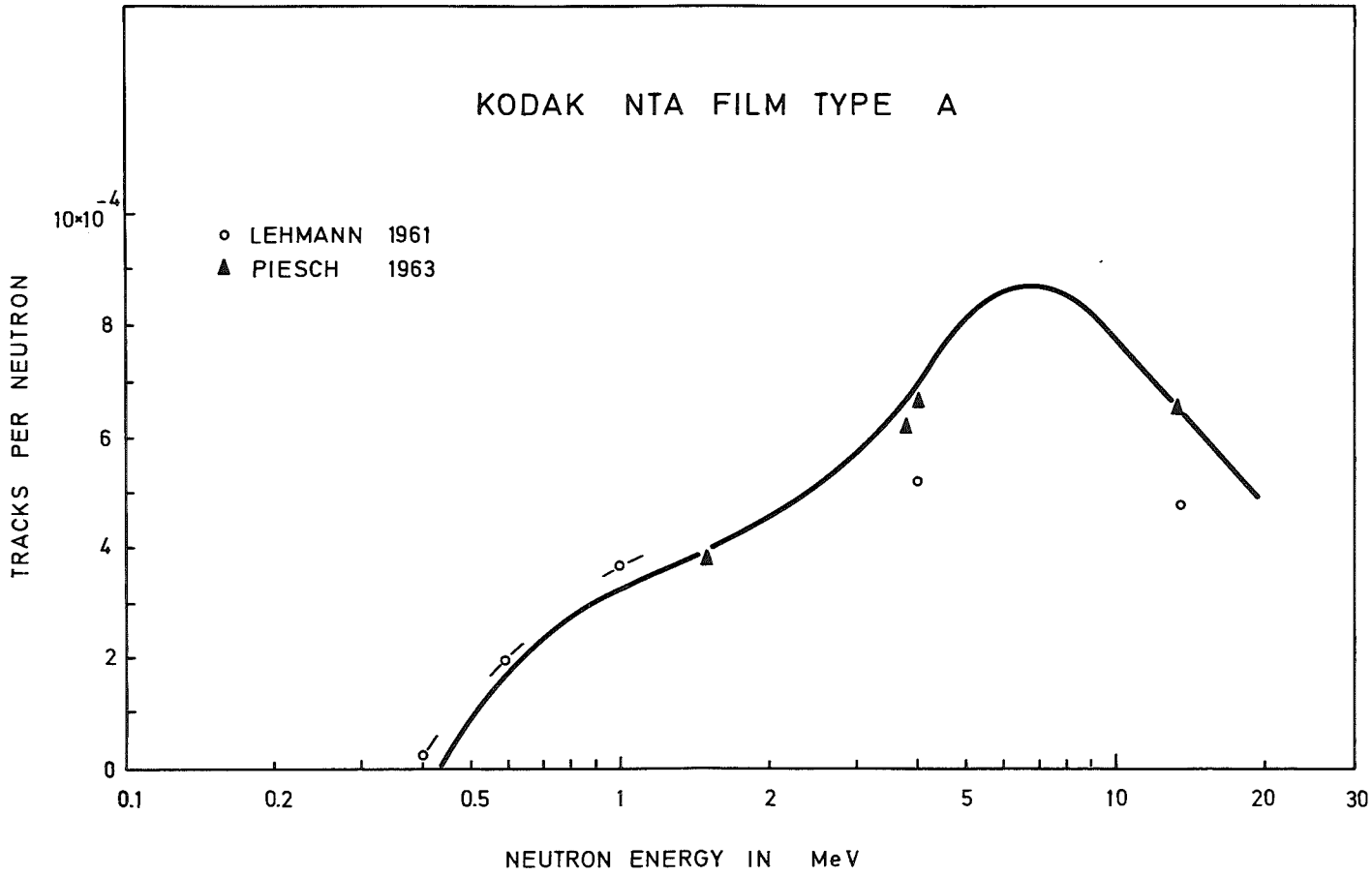

Fig. 1: Energy dependence of Kodak NTA

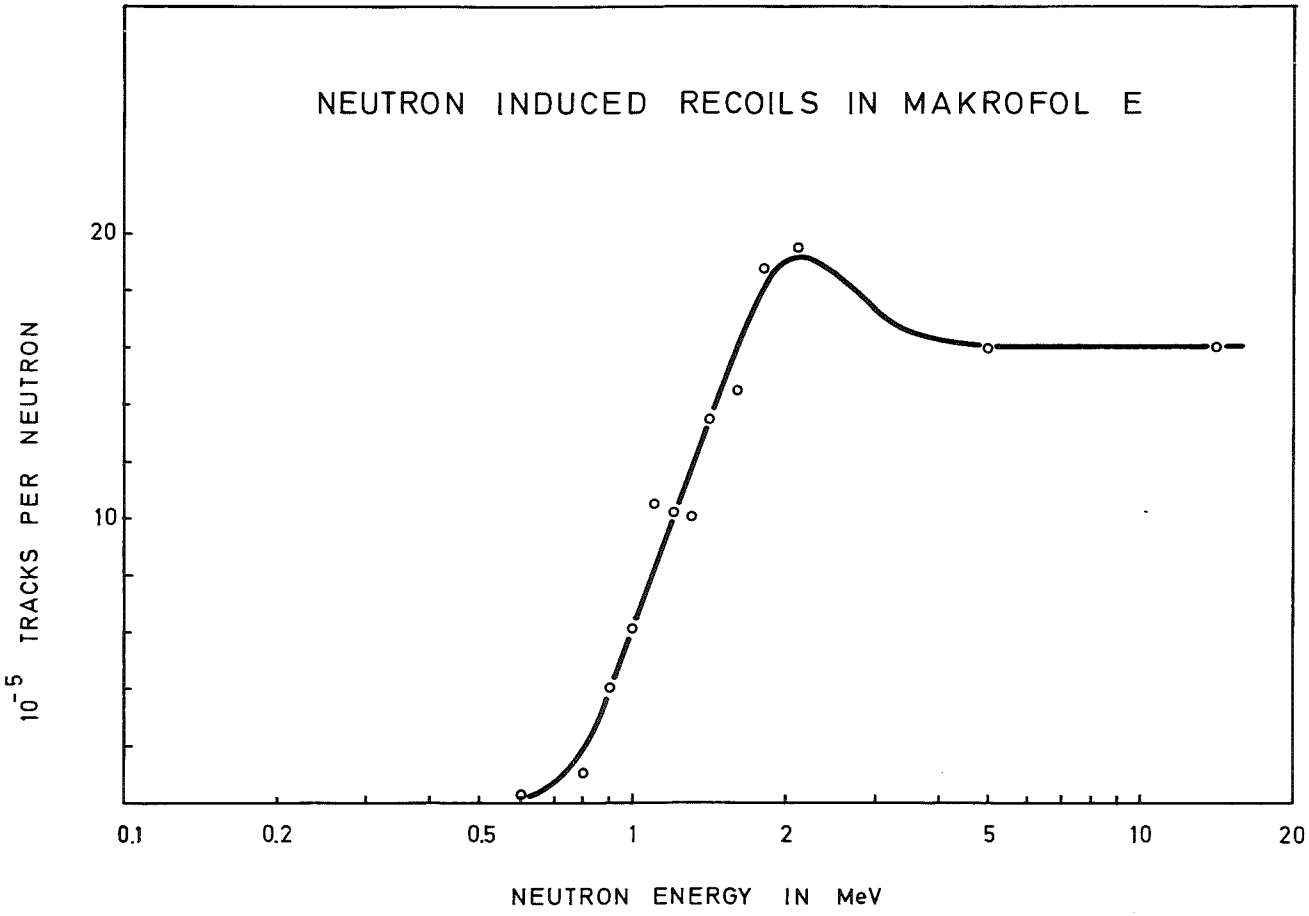

Fig. 2: Energy dependence of neutron induced recoils in Makrofol E

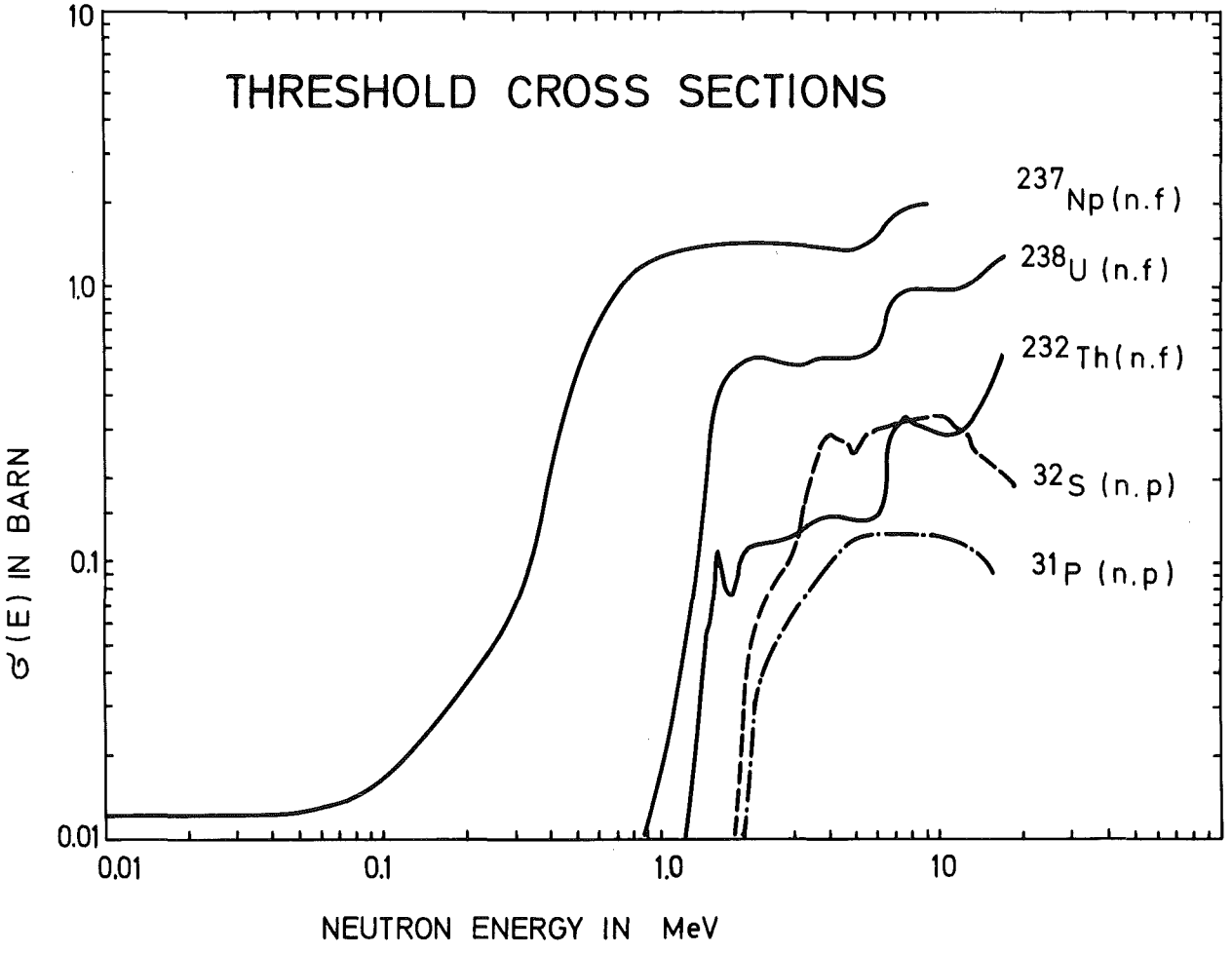

Fig. 3: Energy dependence of neutron cross sections for threshold reactions

humidities used were 0 percent, 53 percent and 97 percent. The humidity was controlled and adjusted to a constant value by means of saturated salt solutions of certain chemicals inside glass jars. These chemicals were silica gel, magnesium nitrate $Mg(NO_3)_2$ and potassium sulphate K_2SO_4. On this basis it was possible to maintain similar relative humidities to those mentioned by Wexler and Hasegawa [18] over the temperatures studied. These relative humidities were 0 %, 53 % and 97 %, respectively.

In order to achieve these humidities it was always important to consider the following steps.

a) The saturated salt solutions had to be enclosed in tightly sealed glass jars
b) The jars, salt solution and the ambient air had to be brought to temperature equilibrium.
c) The salt solution had to occupy as large a surface area as possible
d) The saturated salt solution had to be always agitated before use.

2.3 Irradiation

The samples were irradiated to fission neutrons from a ^{252}Cf source with a neutron emission rate of 1.5×10^9 n/s. The ^{252}Cf source was 10 cm in height and 3 cm in diameter. The dosimeters were irradiated at 40 cm distance from the source. The irradiation time was 2 hours for the NTA film, 16 hours for the Makrofol E and cellulose acetate detectors, and 40 hours for Makrofol with thorium. These irradiation times had been chosen in order to obtain a reasonable number of tracks according to the different sensitivities of the detectors mentioned previously. After exposure, 3 samples from each type of the detectors were developed immediately; i.e., approximately 1 hour after irradiation. The other samples

were placed in the glass jars containing the saturated salt solution. These jars were kept inside a chamber, the air thermostat of which was maintained a constant temperature. For the same temperature, groups of detectors representing the three different relative humidities were stored under these environmental conditions for 1, 2, 8, 10, 12, 20 and 28 days and were then developed at the end of each storage time.

2.4 Processing

2.4.1 Kodak NTA Film

The films were developed for 15 min in a mixture of two compartments A and B:

A		B	
K_2Co_3	50 g	Hydroquinone	0,5 g
$NaSO_3$	10 g		
KBr	5 cm^3	H_2O	500 cm^3
H_2O	500 cm^3		

The films were then placed in a stopbath of potassium bisulphate (40 g per litre) for 5 minutes. They were fixed in Kodak fixer for 2 hours and then washed for 3 hours in running water and finally dried. The developer, fixer, stopbath and washing water were always kept at a constant temperature of 20°C.

2.4.2 Track etching detectors

The different types of detectors were etched as follows:

Material	Etchant composition, temperature and time
Fission fragments in Makrofol KG	35 % KoH, 60°C, 70 min.
Fission fragments in Makrofol E	35 % KoH, 60°C, 80 min.
Recoils in cellulose acetate	26 % NaoH, 60°C, 4 min.
Recoils in Makrofol E	35 % KoH, 60°C, 4 hours

Before etching, all the samples were cleaned with an ultrasonic cleaner and then dried. After etching, the samples also were cleaned and dried before counting.

2.5 Counting conditions

It was demonstrated by Piesch [19] that, by using the new interference contrast technique of Nomarski, an additional contrast can be achieved by producing interference colours in the etch pits different to those in the background of the image. A photo microscope (Carl Zeiss, Oberkochen, Germany) with a Nomarski interference contrast equipment has been used for track counting. For the counting of nuclear tracks the following magnifications were used:

- 12.5 x 20 i.e. 78 x 10^{-4} cm^2/field (Fission fragment)
- 12.5 x 50 i.e. 11.2 x 10^{-4} cm^2/field (Recoils)
- 10 x 80 i.e. 4.52 x 10^{-4} cm^2/field (NTA)

2.6 Accuracy of measurements

The errors involved in the experimental work were mainly due to track counting which caused an error in fading values. The deviation between each set of two samples irradiated under identical conditions and stored at a given temperature and relative humidity was not found to exceed 3 %. The statistical error in counting the number of tracks was σ = 3 %. However, for samples with higher fading results, i.e. high decrease in the number of tracks produced by the effect of temperature and relative humidity, the maximum error was 5-10 %, particularly for Kodak NTA and cellulose acetate. The errors in the dosimeter irradiations were less than 1 %.

3. Results for Kodak NTA Emulsion

3.1 Sealed film results

Figs. 4 - 7 show the relation between the relative number of tracks and storage time at various temperatures and relative

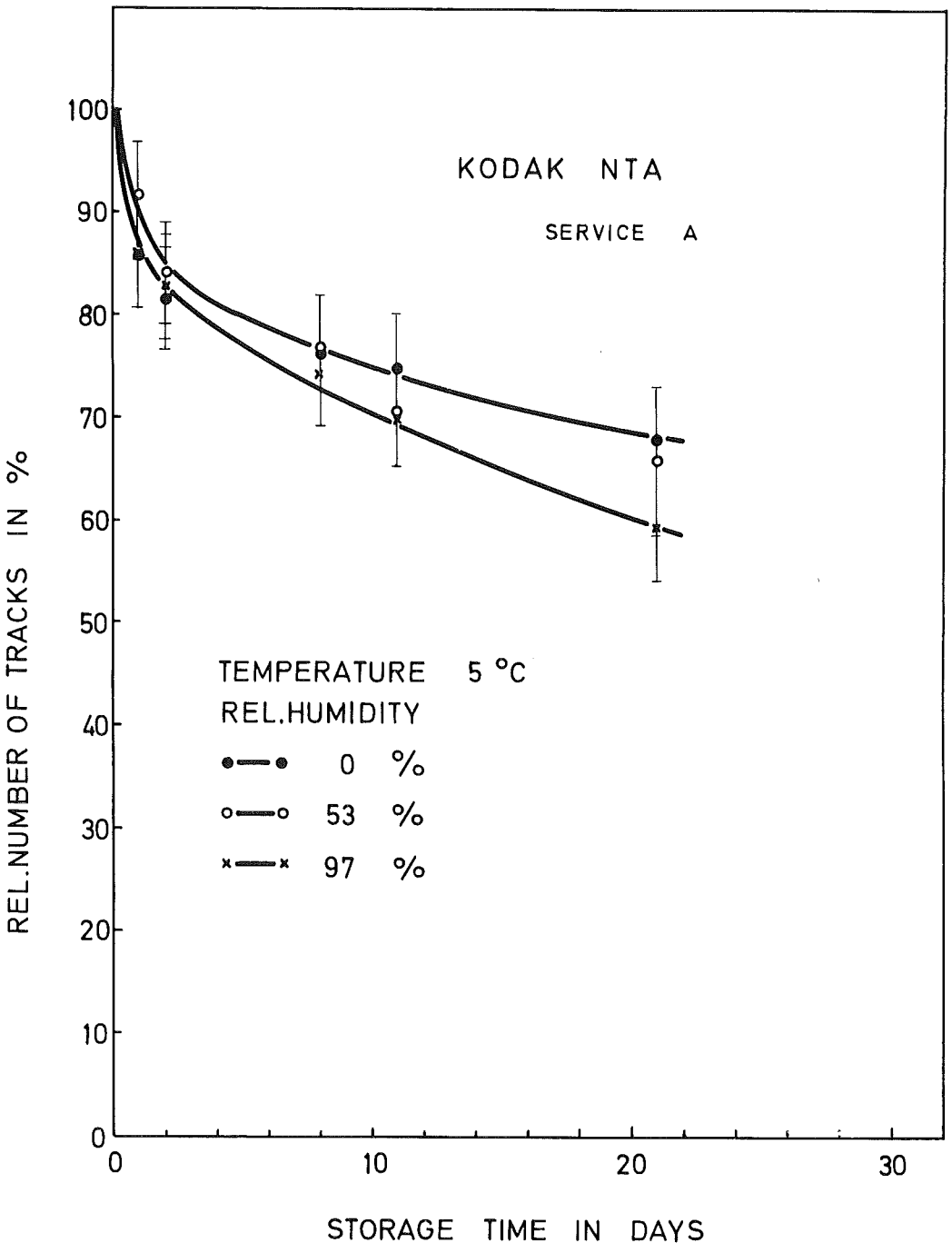

Fig. 4: Variation of the relative number of tracks with storage time for Kodak NTA from service A, stored at 25°C and different relative humidities.

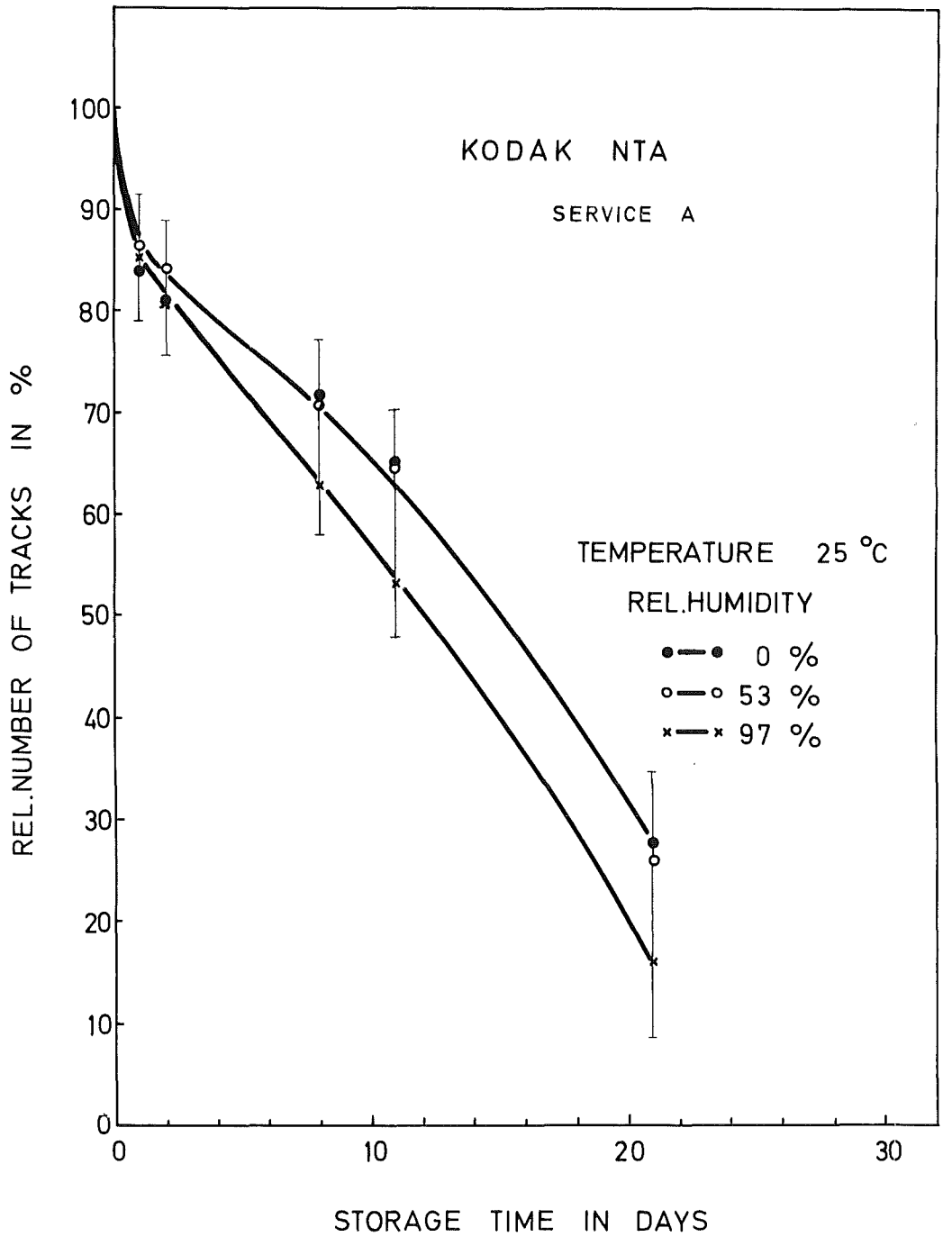

Fig. 5: Relative number of tracks vs storage time for Kodak NTA, service A, stored at 25°C and different relative humidities.

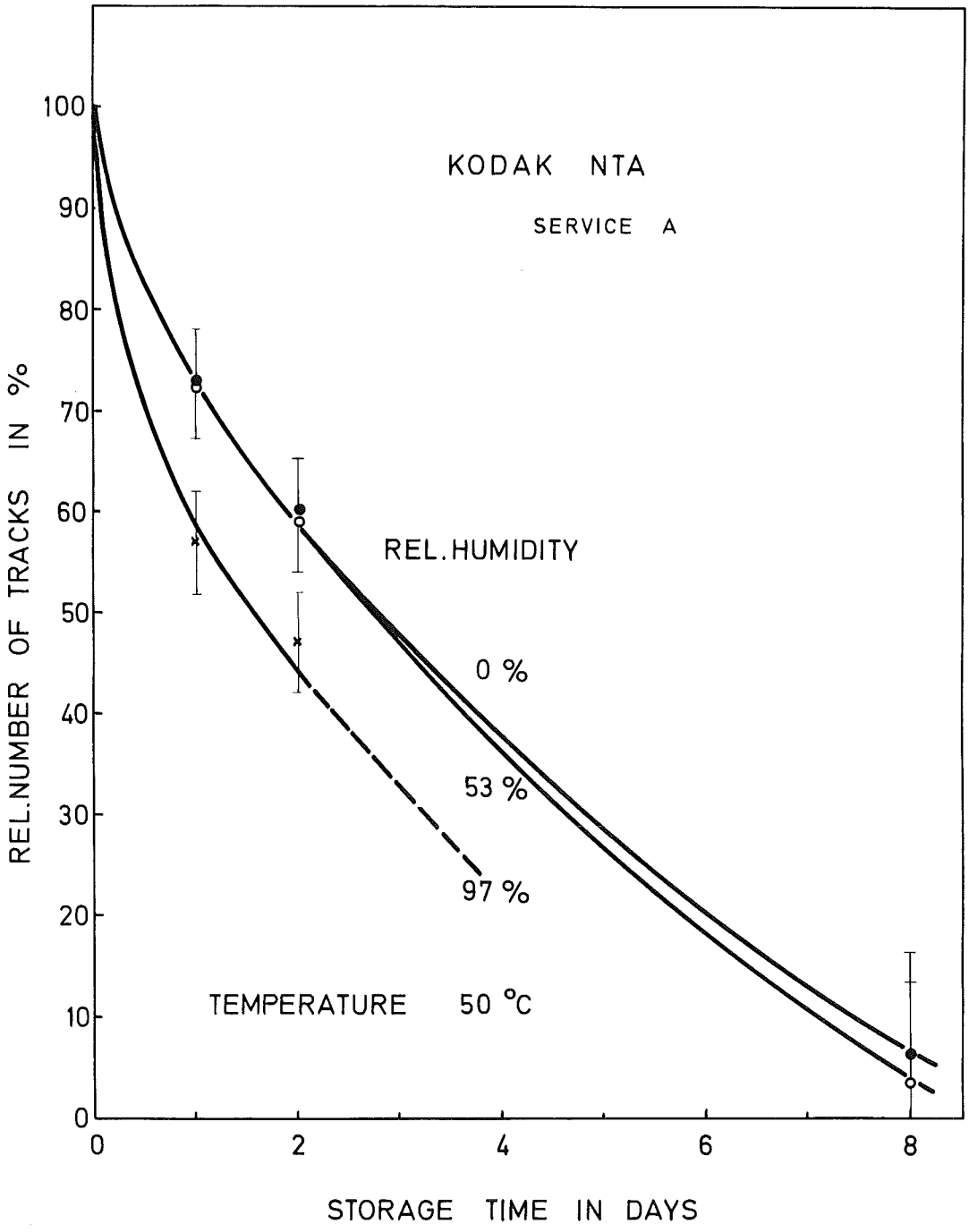

Fig. 6: Relative number of tracks vs. storage time, Kodak NTA, service A, held at 50°C and various relative humidities.

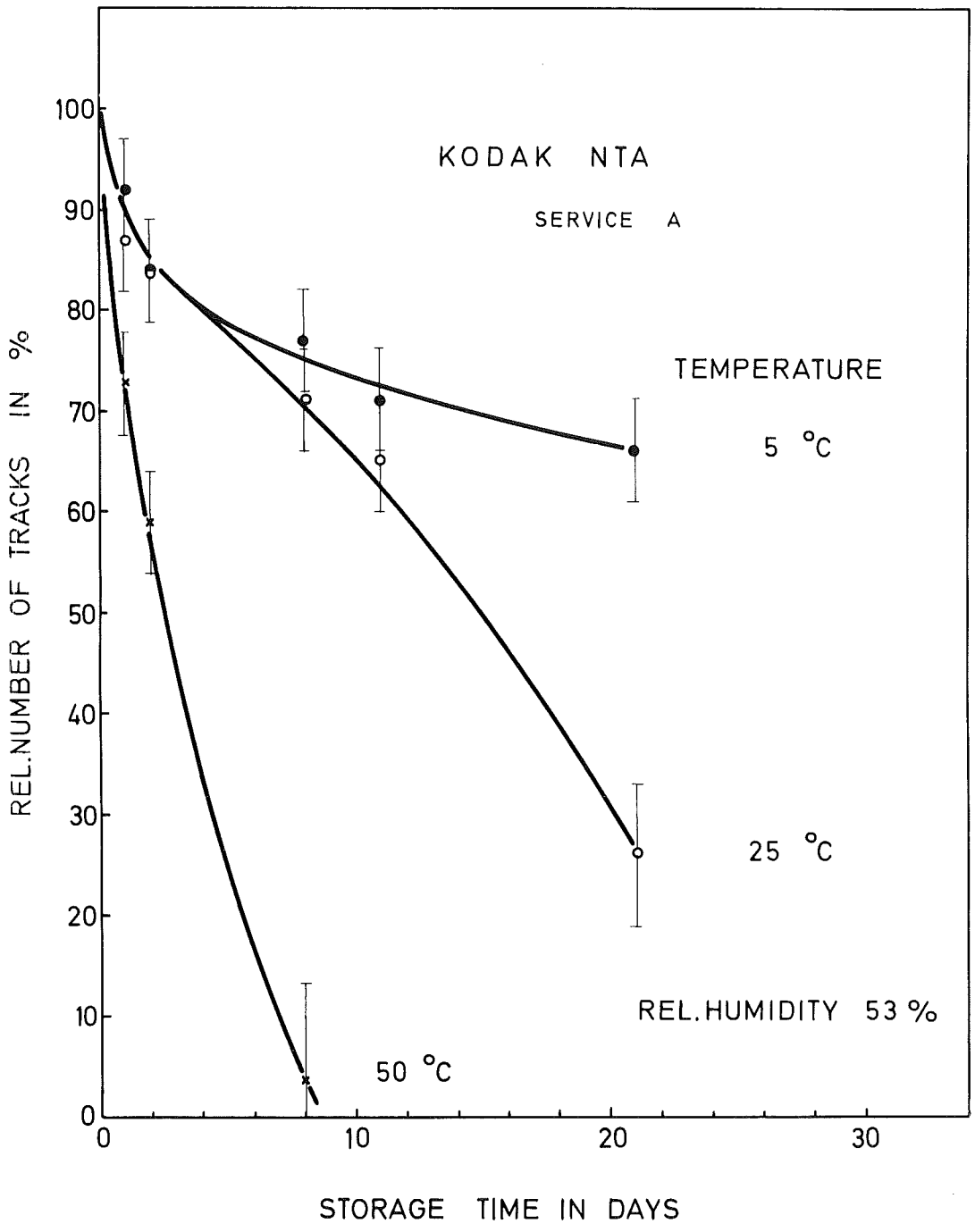

Fig. 7: Kodak NTA from service A stored at 53 % R.H. and different temperatures

humidities. The relative number of tracks is the number of tracks at a given temperature, relative humidity and storage time, divided by the number of tracks of the control films which were developed immediately after irradiation.

It can be shown from the data presented in these figures that, in general, the relative number of tracks decreases strongly when the storage time increases, even for a temperature of 5°C. It can also be observed that there is a strong temperature effect on the latent track fading and the relative humidity does not affect the fading characteristic to such an extent. However, the increase in relative humidity between 53 % and 97 % slightly affects the dependence of the number of tracks on storage time. On the other hand, the increase of temperature from 5°C to 50°C has such a remarkable effect on fading that the relative number of tracks decreases from 80 % to 5 % for a storage time of 8 days at 0 % R.H or 53 % R.H.

Because of the high fading results found at 5°C it is important to consider a relatively high content of humidity in the plastic-aluminium paper in which the NTA film was sealed. Starting from the assumption that the NTA film was sealed under conditions of high relative humidity, we consequently expect an influence of the humidity on fading for relative humidities only which are much higher during the storage period. In our experiments humidity fading was found only for relative humidities above 53 %. It can be seen from experiments reported by Becker [20] that the track density decreases to 78 %, 37 % and 0 % after storage times of 1, 2 or 3 weeks, respectively, of sealed NTA films which were equilibrated for 12 days at 95 % relative humidity and 30°C before sealing. The relative number of tracks found in our experiments at 25°C was 73 %, 55 % and 25 %, respectively.

Actually, the NTA film used in our experiments was found to have been sealed by the personnel monitoring service laboratory A after equilibration under laboratory conditions at 21°C and 50 - 60 % relative humidity. Evidently, a minimum reduction of the number of latent tracks can be obtained only by dessication of the films before additional sealing in the plastic-aluminum paper.

After these first results additional runs were performed with Kodak NTA films by different dosimeter services so that an intercomparison was possible with the following films:

- NTA film (service A) sealed after being equilibrated under laboratory conditions,
- NTA film (service B) sealed with the "normal technique",
- NTA film (service C)* sealed with the "normal technique" in equilibrium with the storage conditions 1 week before the exposure,
- NTA film (service C)* sealed with the "normal technique", no equilibrium with the storage conditions before the exposure, and
- NTA film unsealed i.e. in original wrapping only by the manufacturer.

The results found for the NTA film by the personnel monitoring service B and the service C are given in Figs. 8 and 9. Here the NTA films were stored after exposure at temperatures of 5°C 25°C and 50°C and a relative humidities of 53 % and 97 %, respectively. From these results it is evident that latent track fading is higher for the NTA film from the service B than that from the service C. After a storage time of 2 weeks at 25°C, the residual track density is 45 % for service B and 65 % for service C. A similar track density was found after a storage time of 2 days at 50°C. There is only a slow influence of air humidity on latent track fading. Compared to the results found in the first run, the NTA film from service A shows only a higher fading after a storage time of more than 2 weeks.

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*) NTA films were sealed by Auswertungsstelle für Strahlendosimeter, Gesellschaft für Strahlen- u. Umweltforschung mbH, München

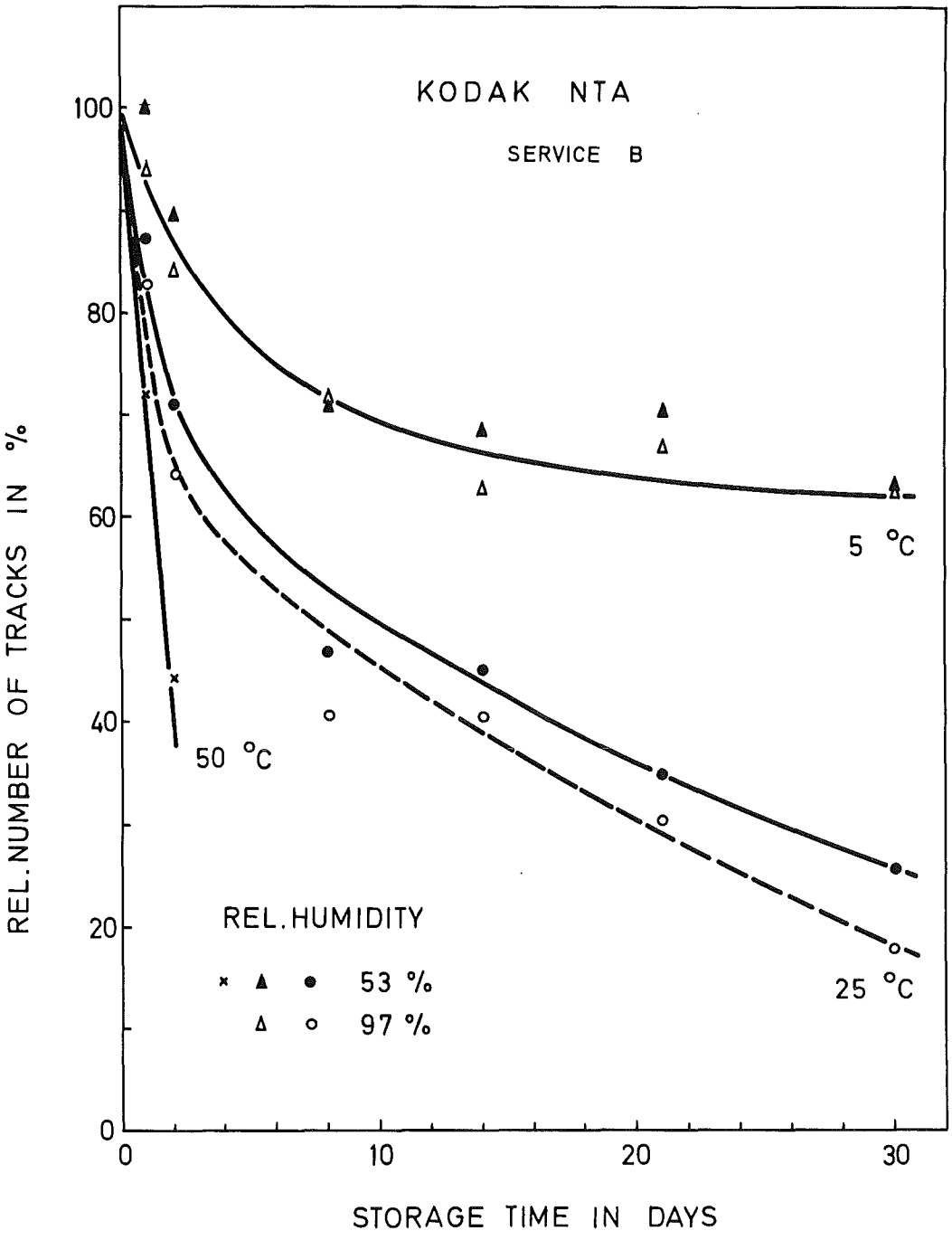

Fig. 8: Relative number of tracks as a function of storage time for Kodak NTA, service B, stored at various temperatures and humidities.

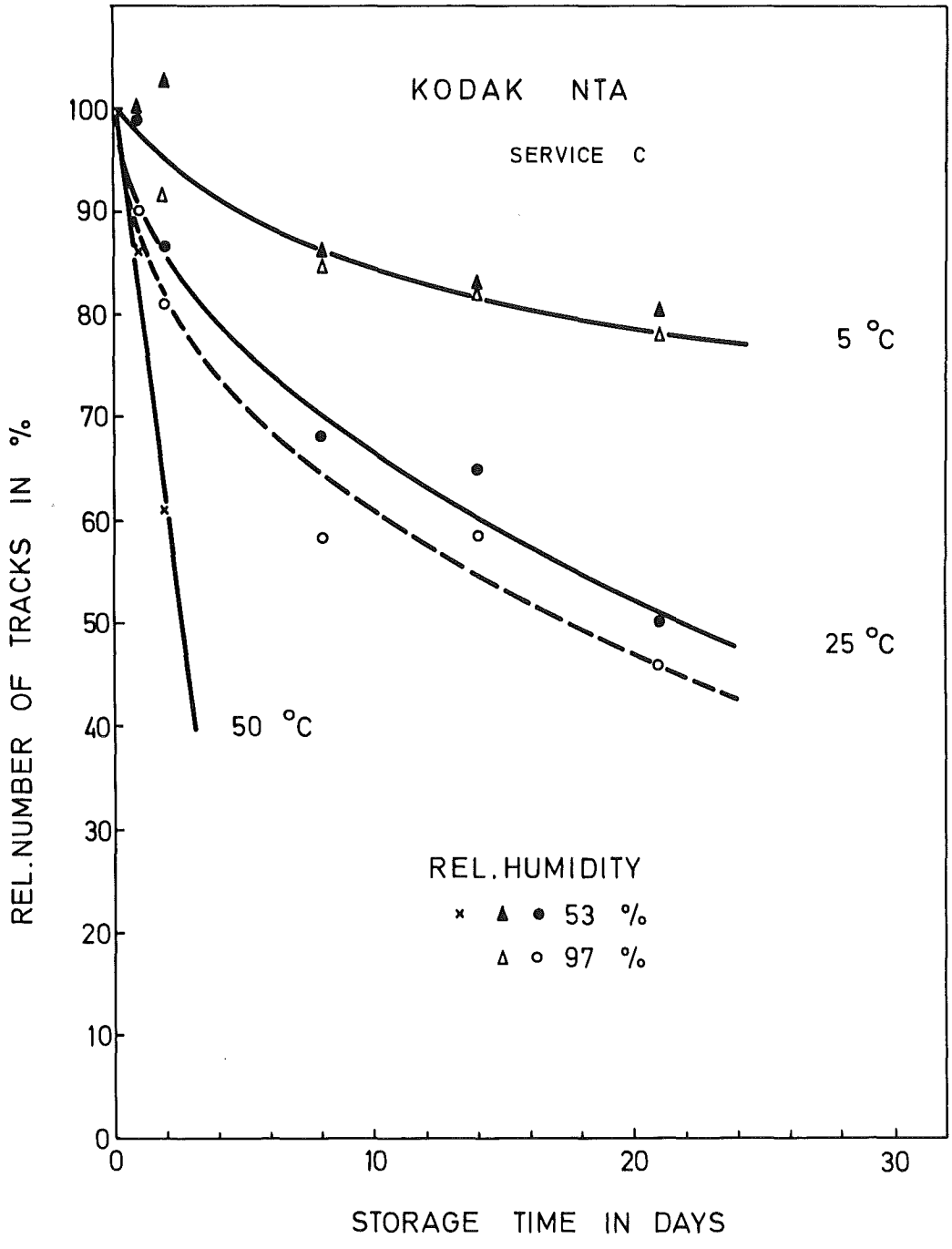

Fig. 9: Variation of the relative number of tracks with storage time for Kodak NTA, service C, for different temperatures and humidities.

If the NTA film is sealed with the "normal technique", there is no significant influence of the storage conditions before the exposure. This is shown in Figs. 10 and 11. The residual track density is plotted vs. the storage time for NTA films from service C, which were equilibrated prior to as well as after the exposure under the same storage conditions and compared to that of NTA films without such equilibrium.

A comparison of the fading results obtained under laboratory conditions is given in Fig. 12 for the NTA films from services A,B,C and for the unsealed film. In the first two weeks after exposure there is a higher fading for the NTA film from service B compared to that found for the unsealed film. The reason for these results can be found in the environmental conditions of the unsealed films which were equilibrated prior to exposure. Therefore, more than 12 days of storage at a relative humidity of 53 % after exposure are necessary to get equilibrium conditions. After this time the influence of fading increases. Contrary to these results, the decrease of the number of tracks is much higher for NTA films from service B, where the sealing process was probably performed under higher humidity conditions or at higher temperatures than for the NTA film from service A, resulting in a higher water content in the sealed film. Under laboratory conditions a residual track density between 25 % and 50 % was found after a storage time of 3 weeks depending on the additional wrapping and the "sealing technique" of the personnel monitoring service, respectively.

In Fig. 13 the relative number of tracks is presented as a function of the temperature during storage for two monitoring services and a storage period of 2 and 14 days, respectively. After a storage period of 14 days at higher temperatures no tracks have been counted. If the NTA film was stored for 2 days at 50°C the residual track density was only in the order of 50 %.

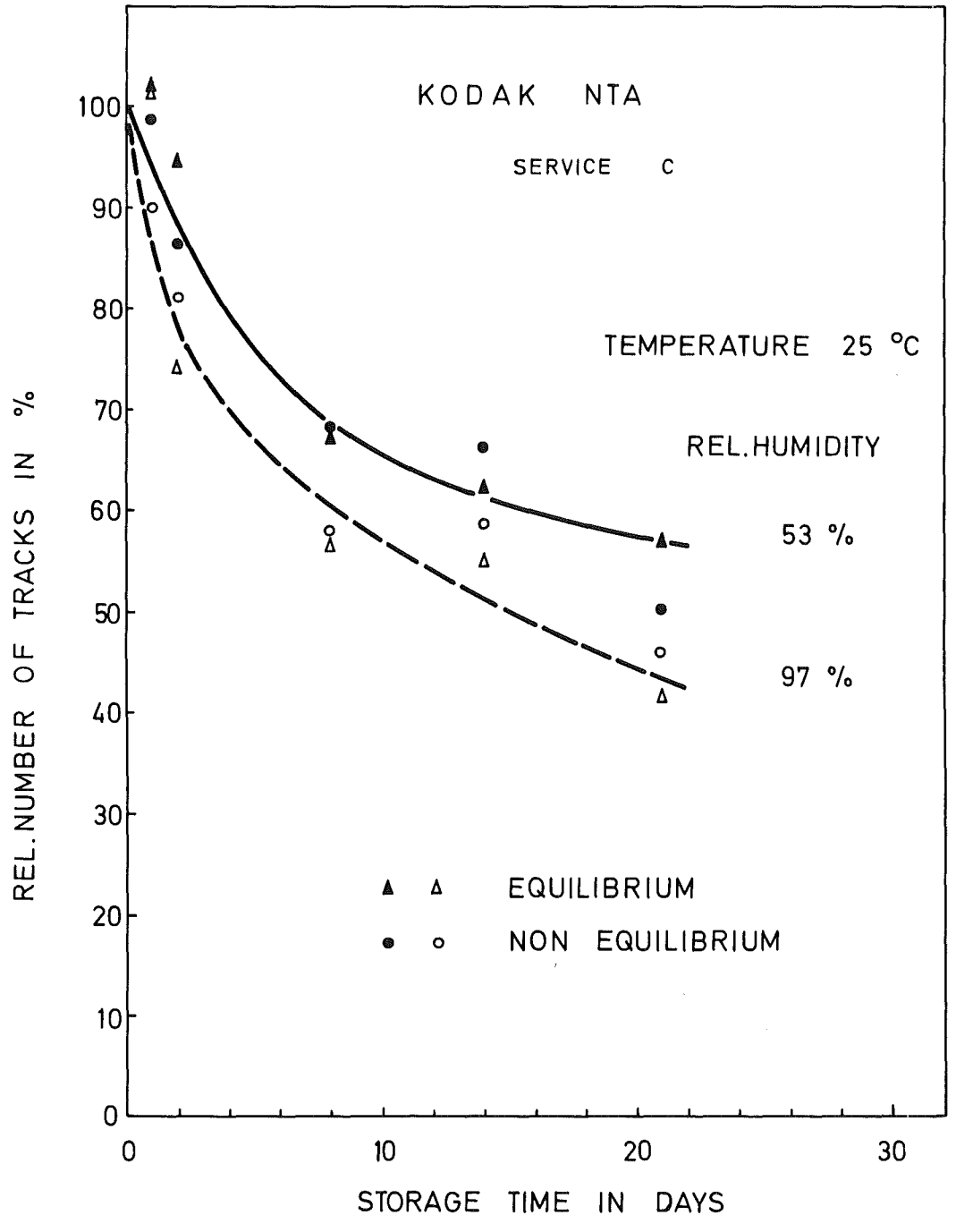

Fig. 10: Latent track fading in Kodak NTA from service C, stored at 25°C and 53 % and 97 % R.H. for films equilibrated for 1 week prior to exposure and for nonequilibrated films

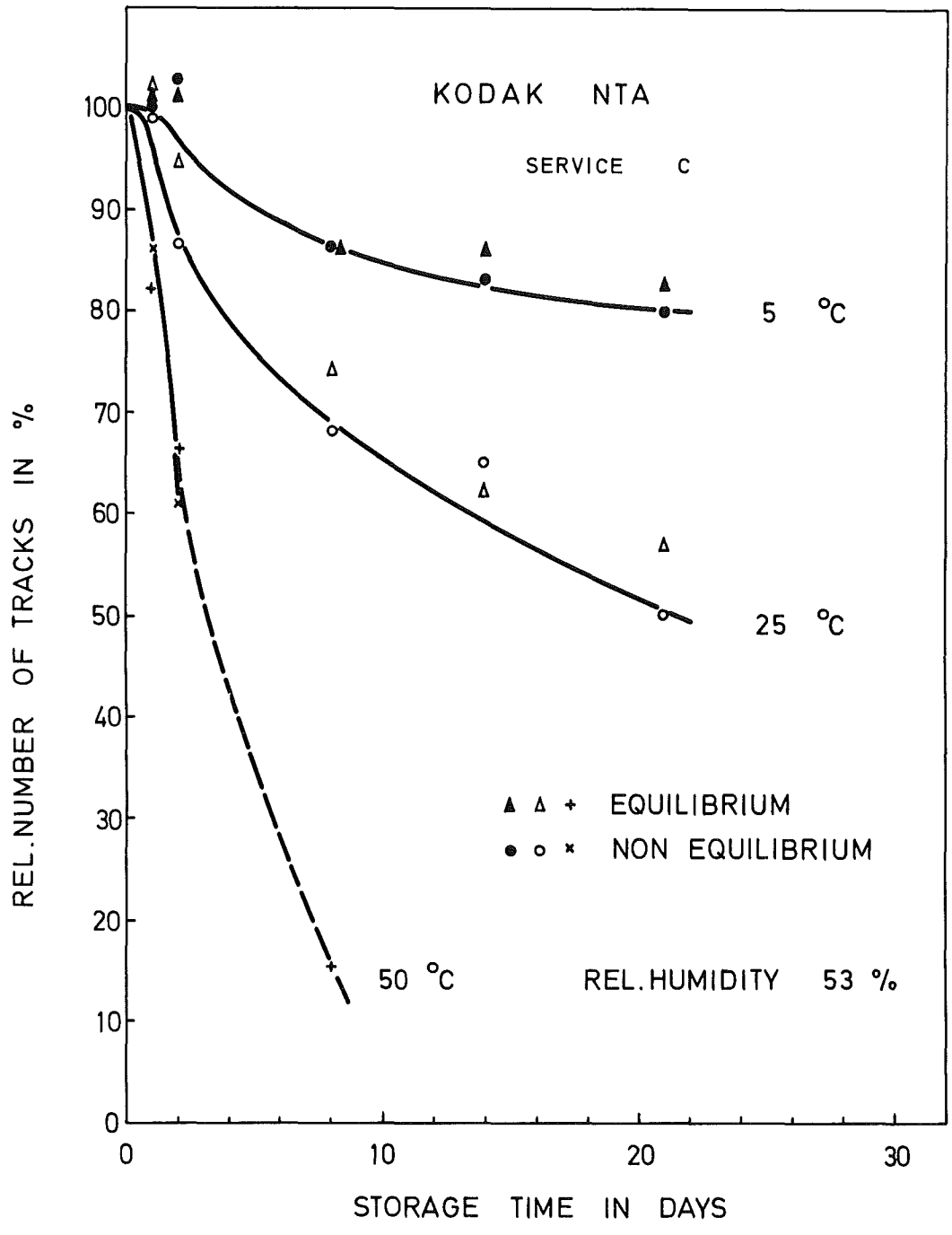

Fig. 11: Fading of latent tracks in Kodak NTA, service C, at 53 % R.H. and different temperatures for 1 week equilibrated and for nonequilibrated films

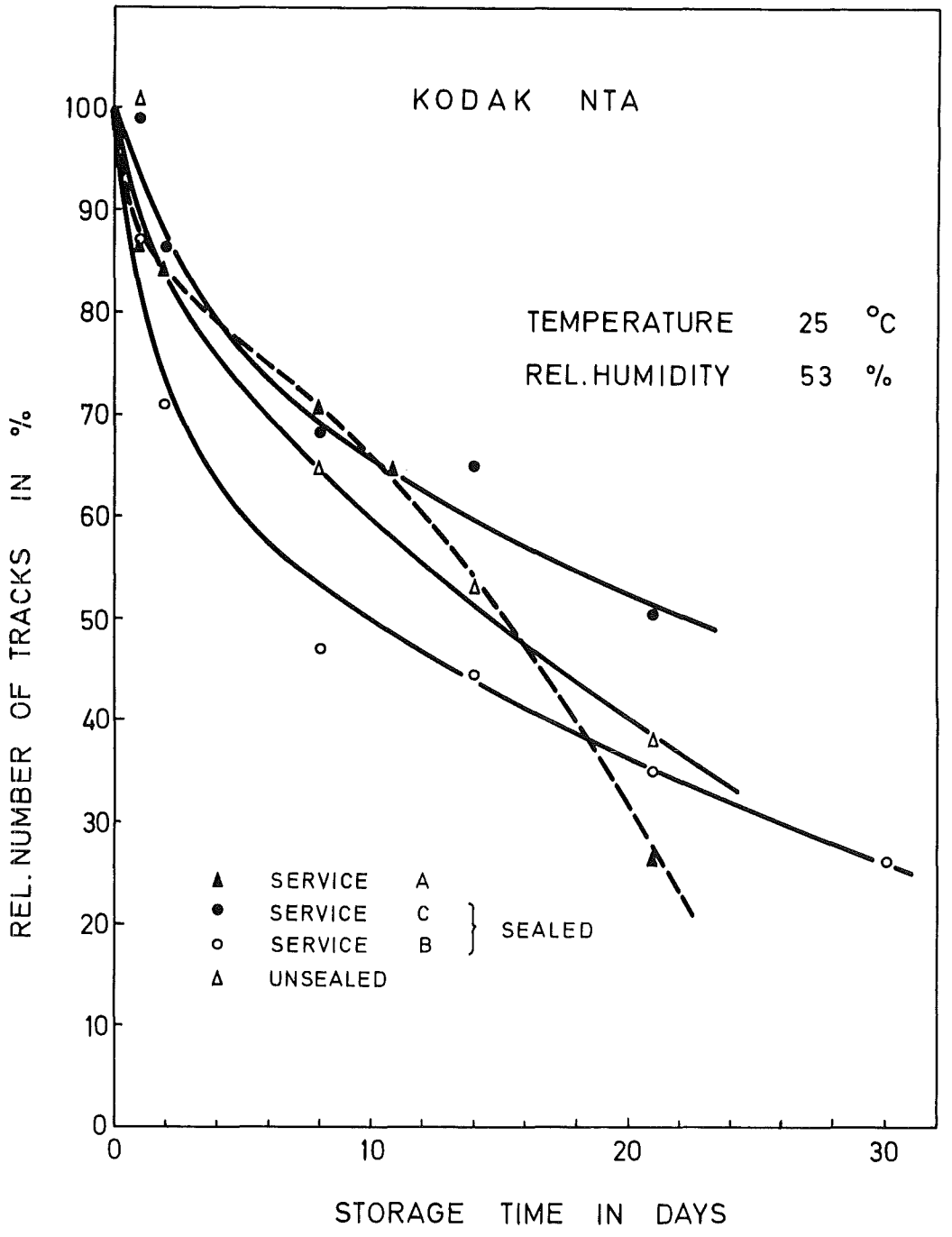

Fig. 12: Comparison of track fading in Kodak NTA from different services for films stored at 25°C and 53 % R.H.

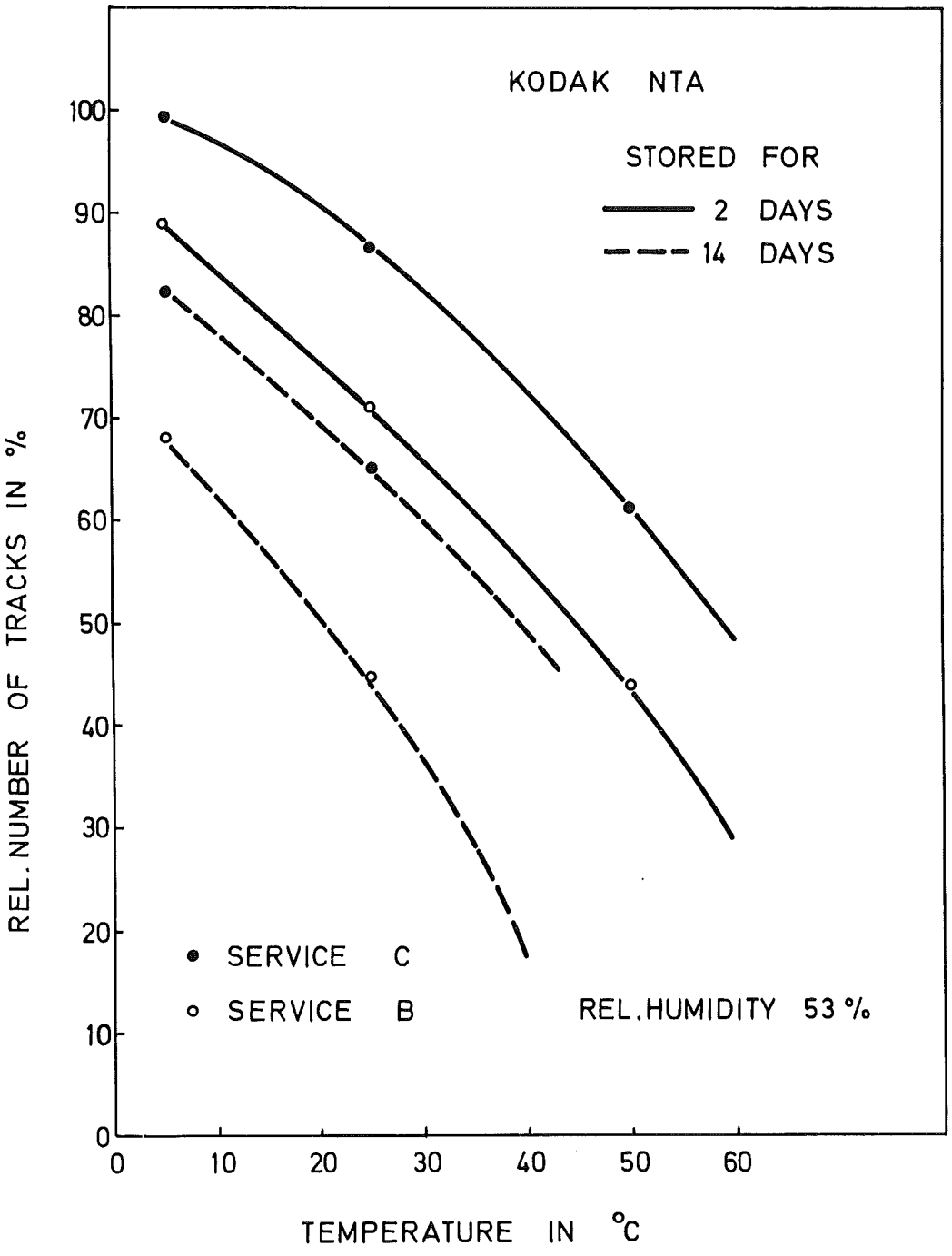

Fig. 13: Relative number of tracks vs. temperature for Kodak NTA from services B and C stored for 2 and 14 days at different relative humidities.

The fading results found for the NTA film from service A, which was equilibrated under laboratory conditions prior to sealing should be finally discussed for short storage periods of 2 days and 8 days, respectively.

Fig. 15 shows the relative number of tracks as a function of relative humidity for Kodak NTA held at temperatures of 5°C, 25°C, 50°C for a period of 2 and 8 days after irradiation. Fig. 14 shows the relationship between the relative number of tracks and temperature for the films stored at 0 %, 53 %, 97 % relative humidity for a period of 2 and 8 days after irradiation. It can be shown Fig. 15 that the increase of relative humidity from 0 % to 97 % does not affect the relative number of tracks for a storage time of 2 days and temperatures of 5°C and 25°C. However, with 8 days of storage time, the increase of relative humidity has a remarkable effect at 25°C. A temperature of 50°C largely affects the relative number of tracks and a remarkable reduction of the number of tracks is shown even for a shorter storage time (2 days).

It can be shown from Fig. 15 that the relative number of tracks decreases from 85 % to 60 % after 2 days and from 75 % to 5 % after 8 days when the temperature increases from 5°C to 50°C at 53 % R.H.

3.2 Comparison with results obtained for unsealed film

The results presented above for Kodak NTA (see also Tab.3) show that high humidities and temperatures may produce fading even when films are stored for 2 days. The experiments reported here show that fading of sealed NTA films may be increased by as much as 50 % at a temperature of 25°C and a relative humidity of 53 % for a relatively short storage period of 10 days after irradiation; on the other hand, the increase of fading is a serious problem at

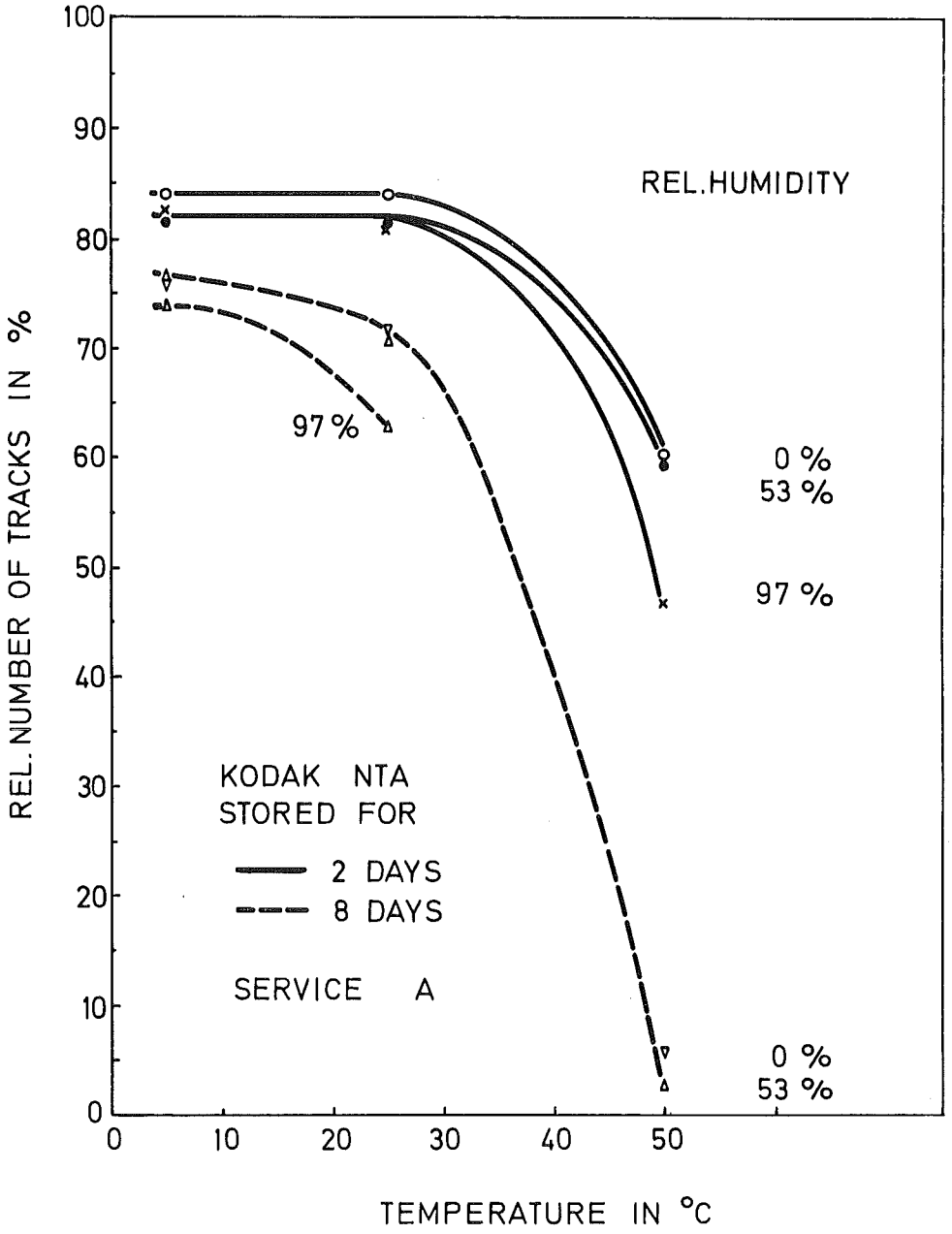

Fig. 14: Latent track fading in Kodak NTA, service A, as a function of temperature, films stored for 2 and 8 days at different relative humidities.

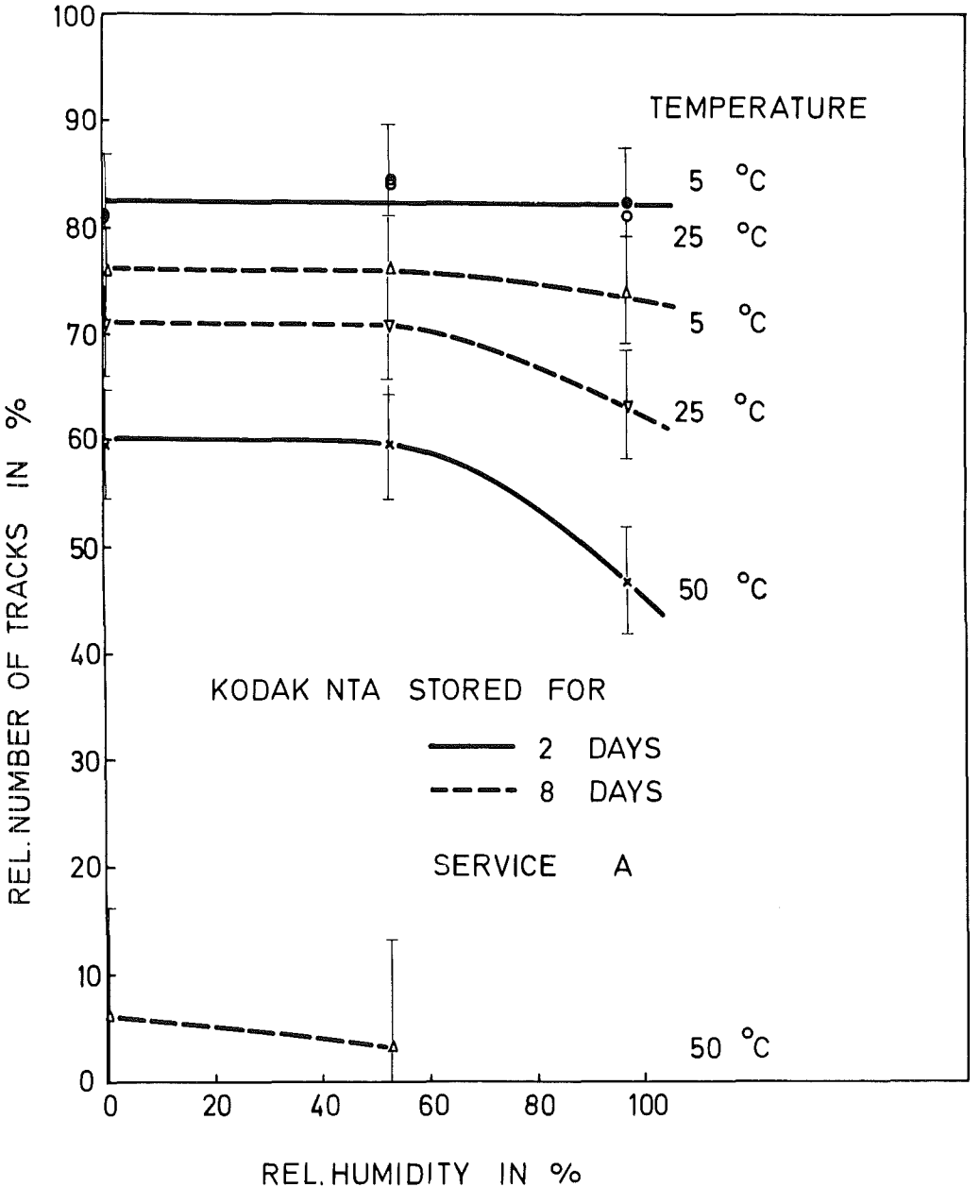

Fig. 15: Latent track fading in NTA, service A, with relative humidity. Films stored at different temperatures for 2 and 8 days.

higher temperatures. For 50°C, fading is ∿95 % after a storage time of 8 days at low humidity. At the same temperature and high relative humidity (97 %), fading amounts to 55 % after 2 days.

The results found with the unsealed NTA film given in Tab. 3 can be compared with the results of other laboratories (see for instance Table 2). At 25°C temperature and 53 % relative humidity 35 % fading was found for an 8 day storage and 47 % for 14 days. No tracks were found after 2 days at 50°C and 97 % relative humidity.

The reported results found with the unsealed NTA film show that the unsealed NTA film should not be used in personnel monitoring even for monitoring periods shorter than one week. At normal temperature and relative humidity (i.e. 24.5°C and 50°C RH), Kahle et al [9] obtained for 0.9 MeV 33 % fading of neutron tracks for a 7 day interval between exposure and development and 56 % for a 14 days interval. Dresel [10] found 100 % fading after 2 days at 55°C and 75 % relative humidity. Becker [20] stated that in a subtropical climate 50 % fading was observed in equilibrated track films without additional wrapping within ∿1.5 days, and ∿90 % of the latent tracks had disappeared after one week.

Jasiak and Musialowicz [22] have noticed that the fading of the unsealed film is 45 % after a storage time of 30 days at 22°C and 60 % relative humidity. The number of tracks decreased to zero after one week at 22°C and 100 % relative humidity. Krishnamoorthy et al [21] found 100 % fading after 10 days for films held at 5°C and 85 % relative humidity. Distenfeld and Klemish [23] stated that 50 % fading was found within 3 weeks at 25°C and 50 % relative humidity.

However, to a certain degree, comparison with other published data is not correct, because a comparison must take into account

also other parameters than temperature and humidity, e.g. the neutron energy, the type and conditions of film sealing, the irradiation period (1 h up to 10 days), the developing conditions, the exact time of developing (the so called "developed immediately after irradiation"), etc.

Seguin [24] reported the results of intercomparison experiments performed by different laboratories of the European Communities. The NTA films were irradiated with neutron energies of 0.7 MeV, 1.5 MeV, 4 MeV and 14 MeV. He stated in the review that the film response experimentally found was lower by a factor 2 for 1.5 MeV neutrons and a factor 6 for 0.5 MeV as compared to the reported one. The fading corrections used (see e.g. Fig.1) by most of the participants are based on experiments performed with Am-Be neutrons with an energy distribution in the range of 4.5 MeV. One explanation for the lower film response may be a different fading influence for lower neutron energies, where the original track length of the microscopically counted recoil protons is shorter and consists of not more than 4 to 6 silver grains. The fading experiments reported by Kahle [9] were performed with 0.9 MeV and 1.3 MeV neutrons. The fading results for the lower energy was higher by a factor 2 (see also Table 2).

The response of the Kodak NTA film which was developed immediatly after the exposure to ^{252}Cf fission neutrons (E $\sim$1.9 MeV) was found to be 4 x 10^{-4} tracks/neutron and was in good agreement with previous calculations and experiments (see Fig.1).

From the results presented in this part, we can conclude that the Kodak NTA film is not recommended as a personnal dosimeter for a long period of use i.e. for more than 14 days. Even under normal environmental conditions, remarkable fading can be expected. However, when it is necessary to use this film, attention must be paid to the effects of environmental conditions and correction factors found experimentally must be considered. Another solution is to expose calibration films to the same environmental conditions under which the monitoring films are actually used

Table 3: Residual tracks in Kodak NTA after a storage at various environmental conditions

service	storage time (days)	RELATIVE NUMBER OF TRACKS %					
		5°C		25°C		50°C	
		53 % R.H.	97 % R.H.	53 % R.H.	97 % R.H.	53 % R.H.	97 % R.H.
A	1	91.7	85.8	86.6	85.3	72.8	57.3
	2	84.3	82.5	84.3	81.1	59.4	46.8
	8	76.7	74.3	71.2	63.3	3.5	zero
	14	72	66	54	44	zero	zero
	21	65.9	59	26	16.4	zero	zero
B	1	100	94	87.3	82.7	71.5	64.4
	2	89.4	84	71	63.8	43.8	33.3
	8	70.5	71.5	47	40.5	zero	zero
	14	68.5	62.4	44.7	40.6	zero	zero
	21	71.5	67	34.9	30	zero	zero
	30	63	62.9	25.6	17.8	zero	zero
C	1	100	--	99	90.3	86	77
	2	103	91.3	86.5	81	61	54.6
	8	86.2	84.7	68.2	58	zero	zero
	14	83.2	82.3	65	58.8	zero	zero
	21	80	78.5	50	45.8	zero	zero
C*	1	101	102	102	102	81.8	65.4
	2	101.5	101	94.5	74.1	66.4	49.7
	8	86.2	85.1	74	56.4	15.4	zero
	14	86	85.7	62.2	54.6	zero	zero
	21	82.5	73	57	40.8	zero	zero
unsealed	1	100	90.3	101	--	91.5	zero
	2	1o5	75	101	zero	zero	zero
	8	83.4	zero	65	zero	zero	zero
	14	78.5	zero	53.3	zero	zero	zero
	21	68.3	zero	48.3	zero	zero	zero

*) equilibrated

for approximately the same period. For extreme climatic conditions in a subtropical climate even the aluminium - plastic sealed NTA film is not an adequate detector for personnel monitoring and cannot be recommended for a period of more than 2 days.

4. Results for fission fragment detection

4.1 In Makrofol E

The relative number of tracks as a function of storage time are illustrated in Figs. 16 and 17 for different temperatures at a relative humidity of 97 % and for different relative humidities at a temperature of 50°C. It can be noticed from the data presented in these figures that there is practically no fading at temperatures below 50°C even for a storage time of 4 weeks. At 25°C and 97 % R.H., fading after 4 weeks was found to be 4 % which is within the statistical error. At 50°C, fading started to be remarkable and was found to be dependent on the relative humidity and storage time as shown in Fig. 17. At this high temperature a loss of 8 % was observed after 4 weeks at 53 % R.H. As the relative humidity increased to 97 %, 18 % fading was observed under these extreme conditions of temperature and relative humidity.

4.2 In Makrofol KG

Figs. 18 and 19 show the relative number of tracks as a function of storage time at various temperatures and relative humidities for 97 % R.H. and 50°C, respectively. Fig. 20 shows the relative number of tracks as a function of temperature at different relative humidities. The results presented in these figures show that the fading characteristics are similar to that in Makrofol E; At 50°C and 97 % R.H. 20 % fading was observed after 4 weeks instead of 18 % fading for Makrofol E.

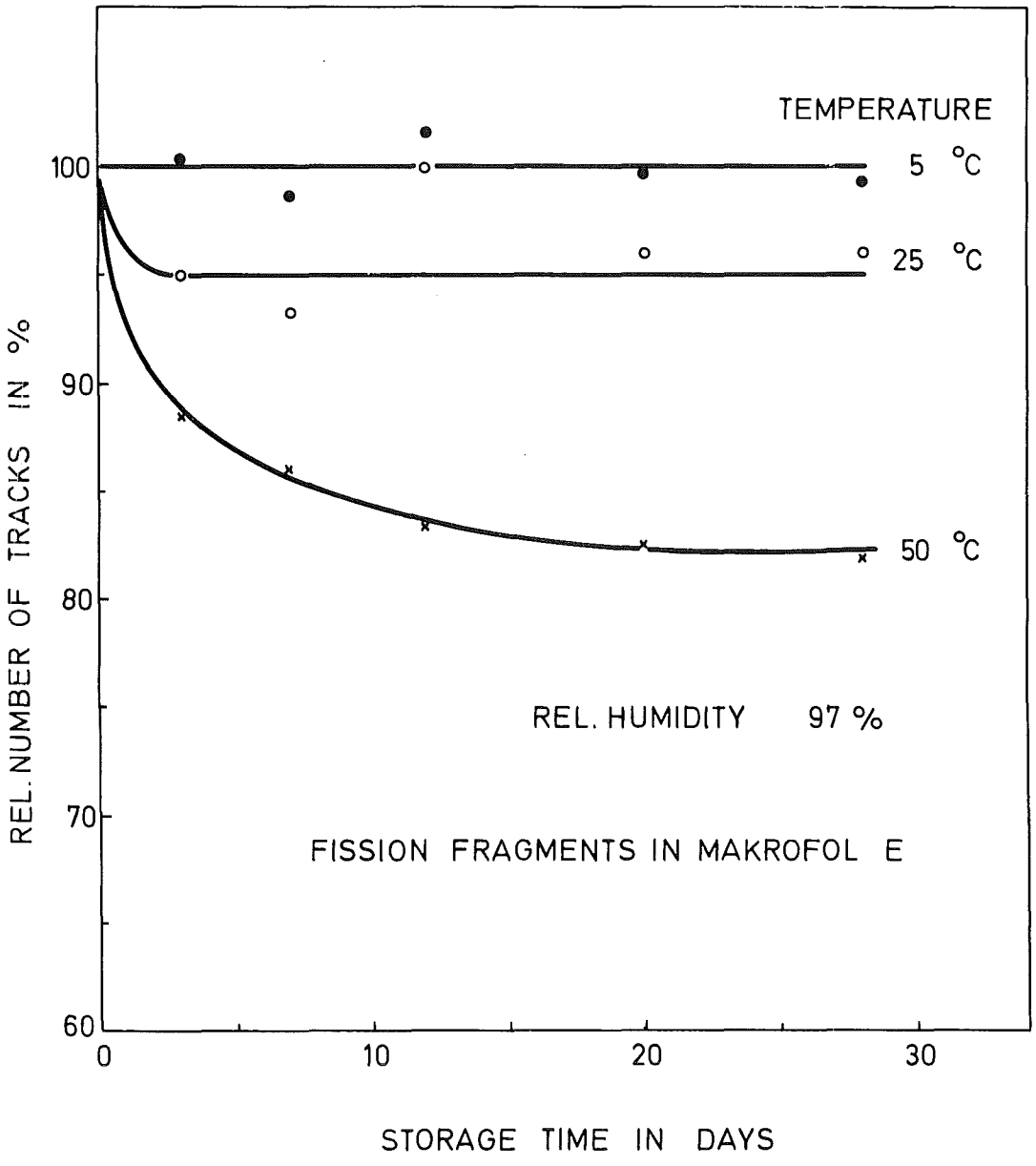

Fig. 16: Relative number of track as a function of storage time for fission fragments in Makrofol E stored at 97 % R.H. and different temperatures

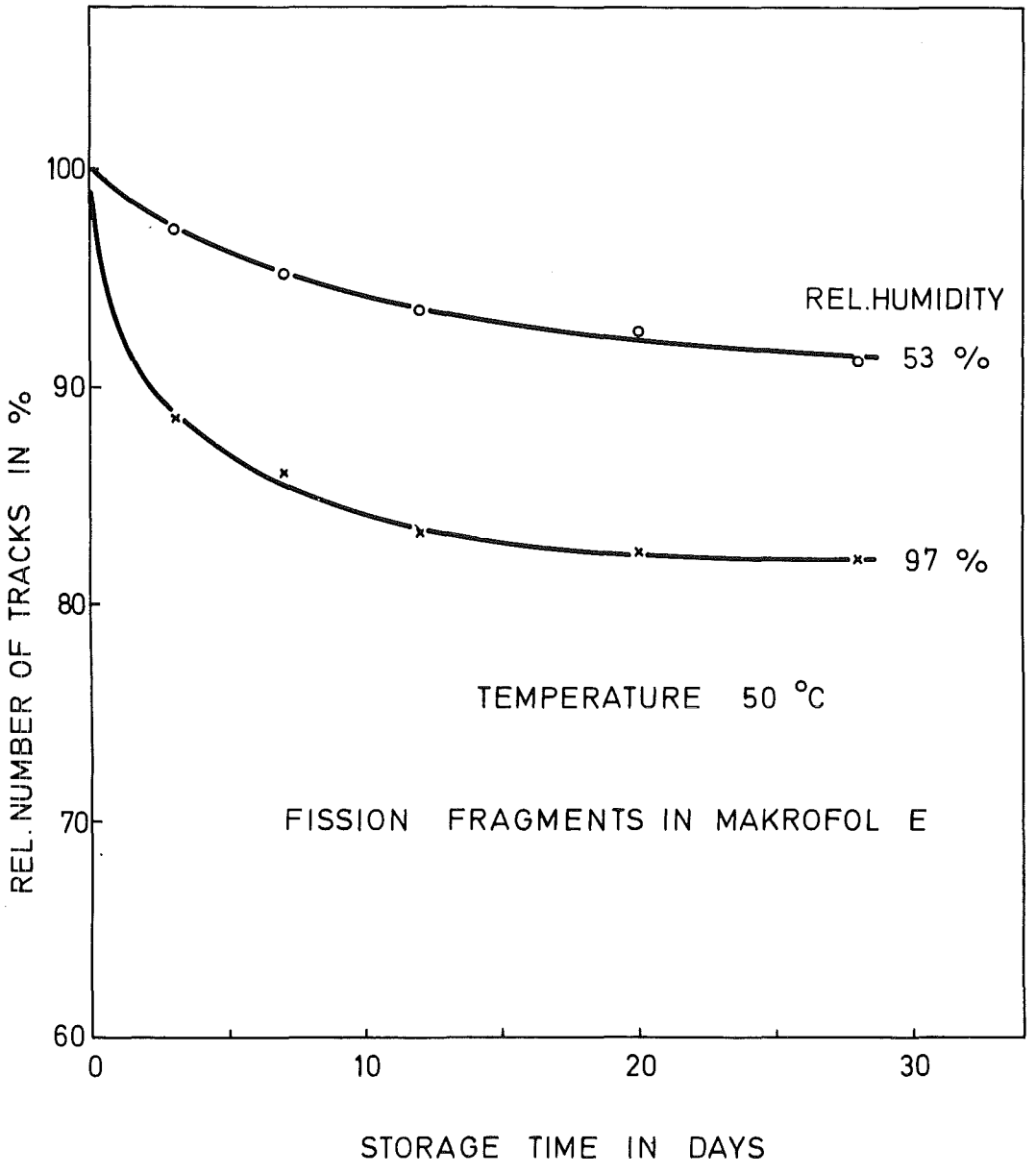

Fig. 17: Relative number of track as a function of storage time for fission fragments in Makrofol E stored at 50°C and different relative humidities.

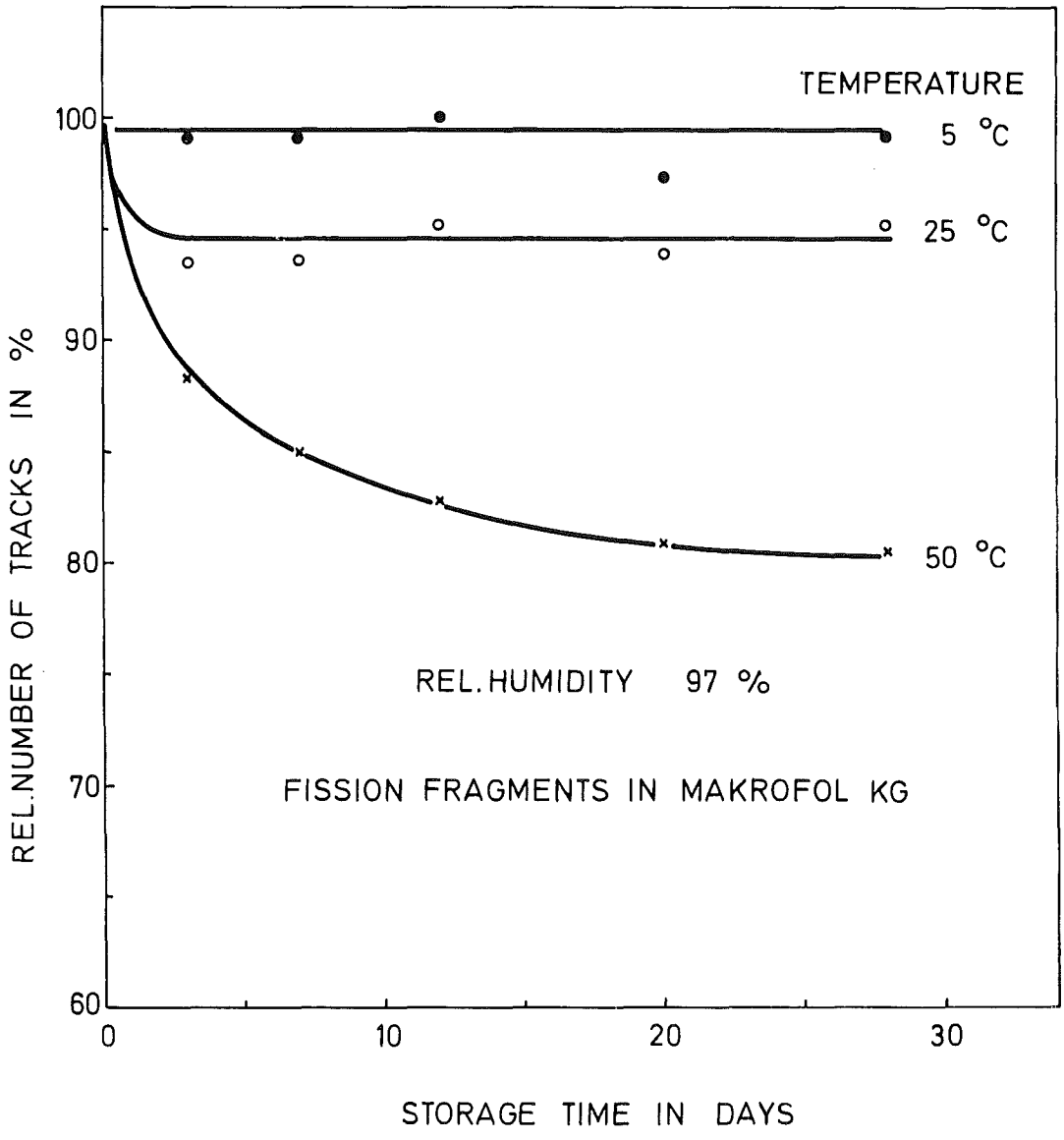

Fig. 18: Latent track fading in Makrofol KG stored at 97 % R.H. and different temperatures

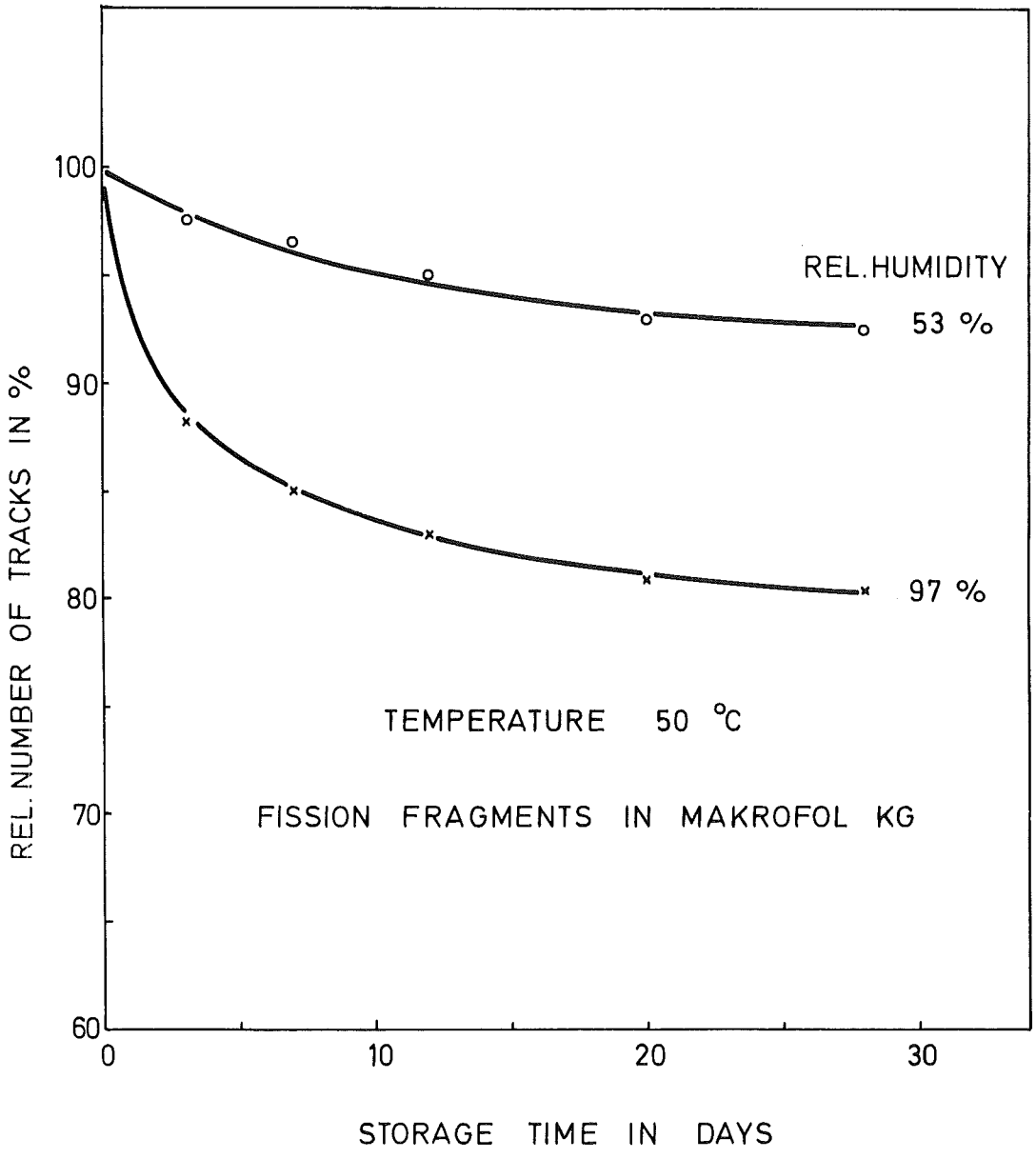

Fig. 19: Latent track fading in Makrofol KG held at 50°C and different relative humidities.

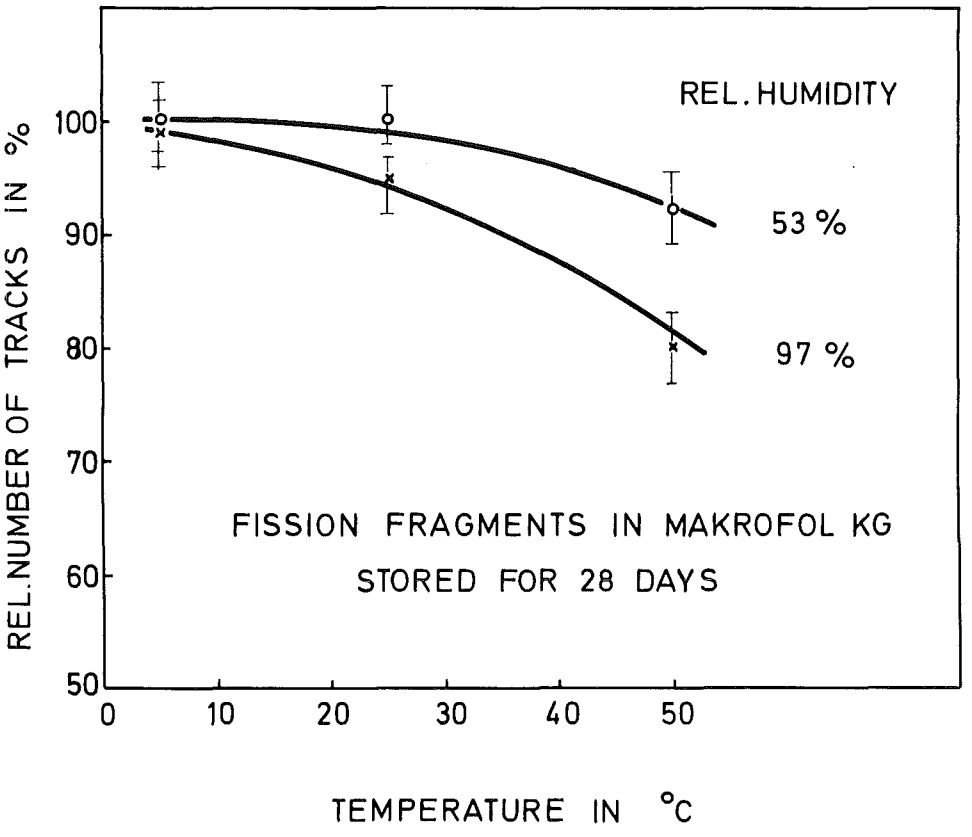

Fig. 20: Relative number of tracks vs. temperatures for fission fragments in Makrofol KG stored for 28 days at different relative humidities.

In general, the data presented in this part showed that, for fission fragments in Makrofol (E and KG), no fading of the latent fission fragment tracks has been observed at room temperature. It also showed that fission fragments in Makrofol are stable for at least 4 weeks at 25°C and 97 % relative humidity. Fading is not remarkable at temperatures below 50°C, but is slightly affected by higher relative humidities.

These results are in agreement with those obtained by Becker [20], who found 25 % fading after 3 weeks at 60°C and 95 % R.H. and spark counting for fission fragments in Kimofol 10µm. At a temperature of 120°C the latent fission fragment tracks in Makrofol E show 25 % fading after 1 hour of storage.

5. Results for neutron induced recoils and α-particles

5.1 In cellulose acetate

Figs. 21 and 22 show the relative number of tracks as a function of storage time for constant temperatures of 25°C and 50°C and different relative humidities. Figs. 23 and 24 illustrate the same relationship for constant humidities of 53 % and 97 % R.H. and different temperatures.

It can be seen from these figures that at 0 % R.H., the relative number of tracks is relatively independent of the storage time even at higher temperatures. The increase in the relative humidity causes high fading dependent on the storage time. At 25°C and 97 % relative humidity, a 50 % reduction of the relative number of tracks was observed after one week, as shown in Fig. 21. If the temperature increases, the dependence of the relative number of tracks on storage time is very high, particularly at higher relative humidities. For 50°C and 97 % R.H. the tracks disappeared completely after about 10 days, as shown in Fig. 22. Moreover, for these extreme conditions of temperature and relative humidity a damage effect in the surface structure of cellulose acetate has been noticed, as shown in Fig. 23.

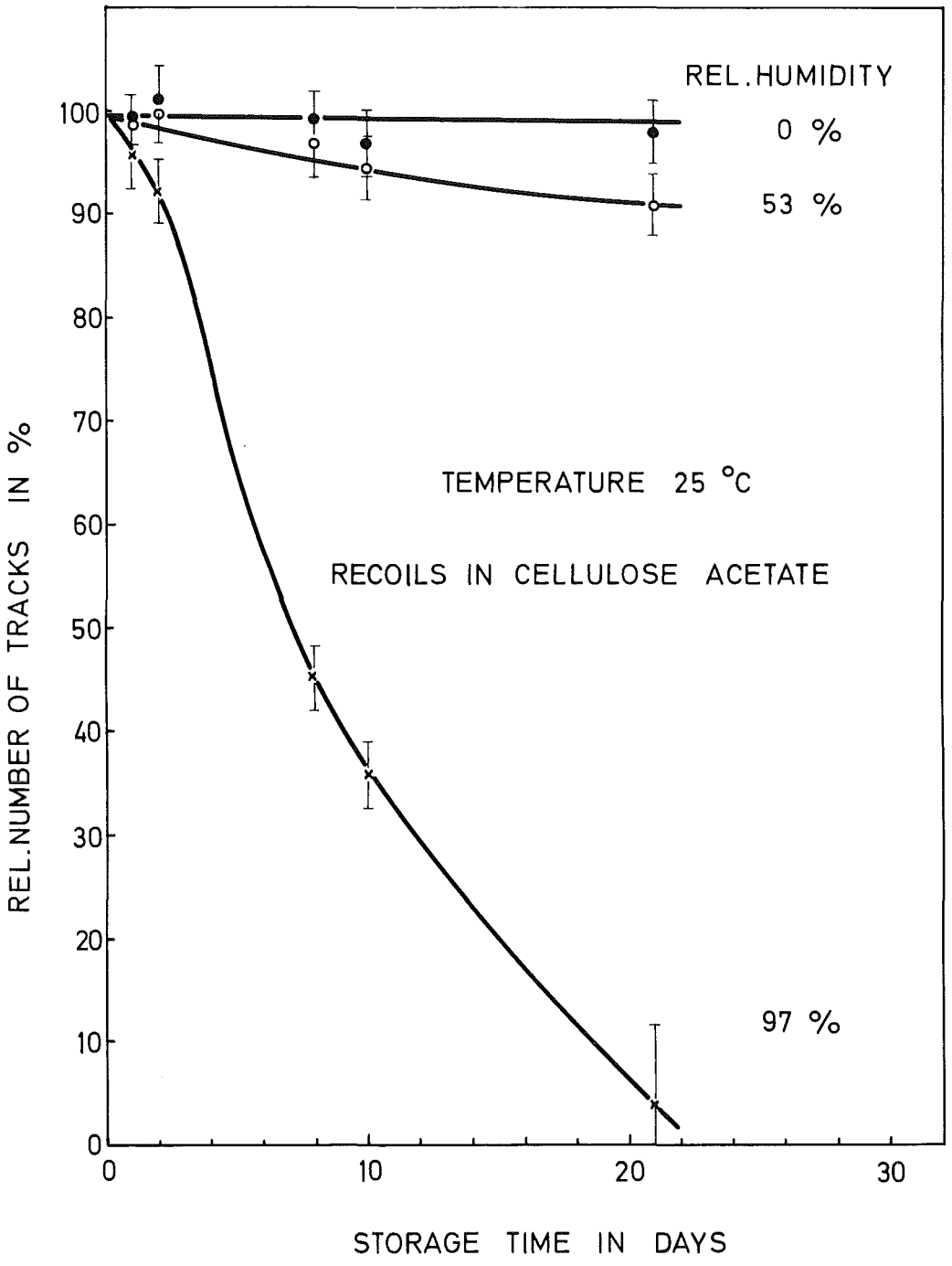

Fig. 21: Relative number of tracks as a function of storage time for neutron-induced recoils in cellulose acetate at 25°C and different relative humidities.

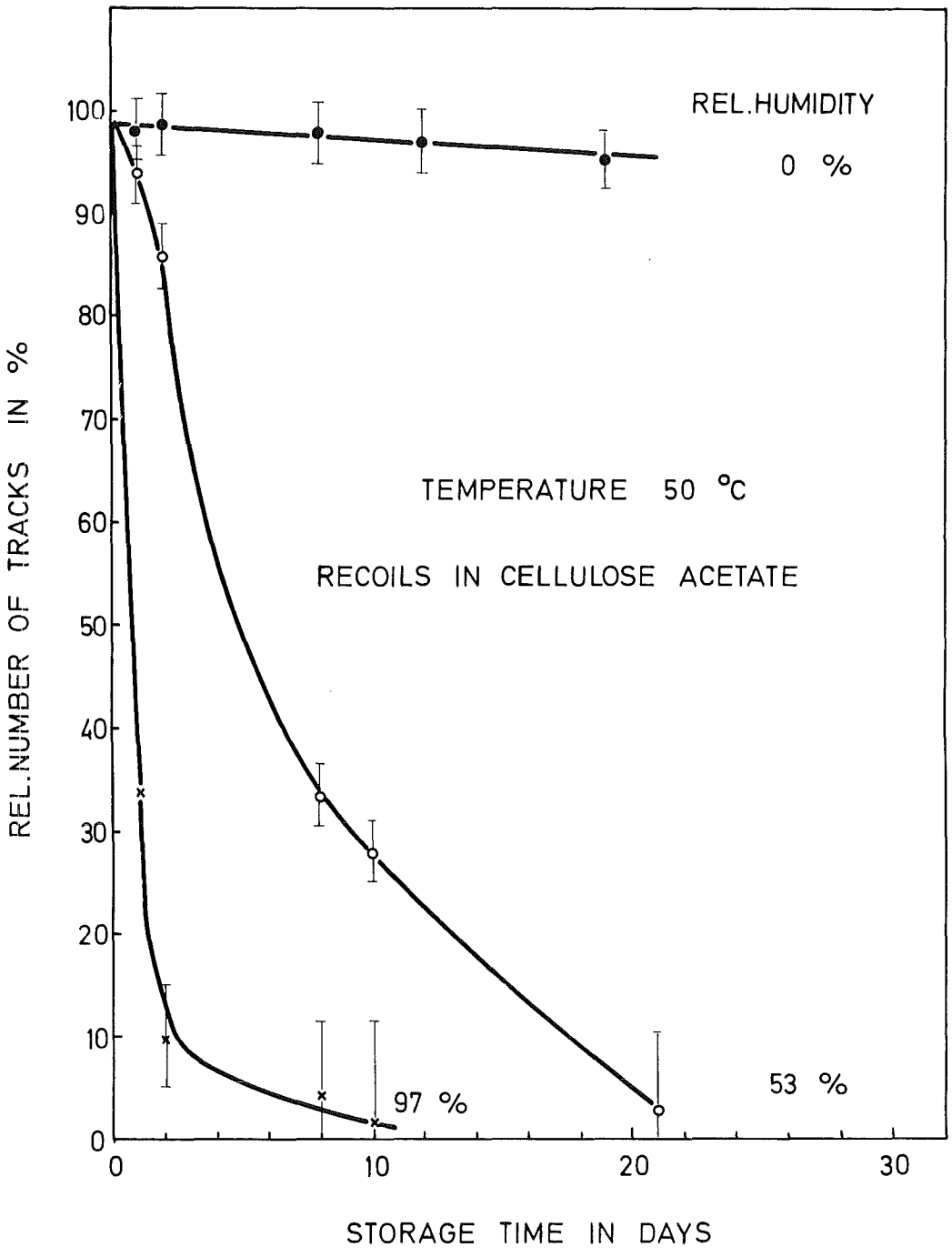

Fig. 22: Latent track fading for neutron induced recoils in cellulose acetate at 50°C and various relative humidities.

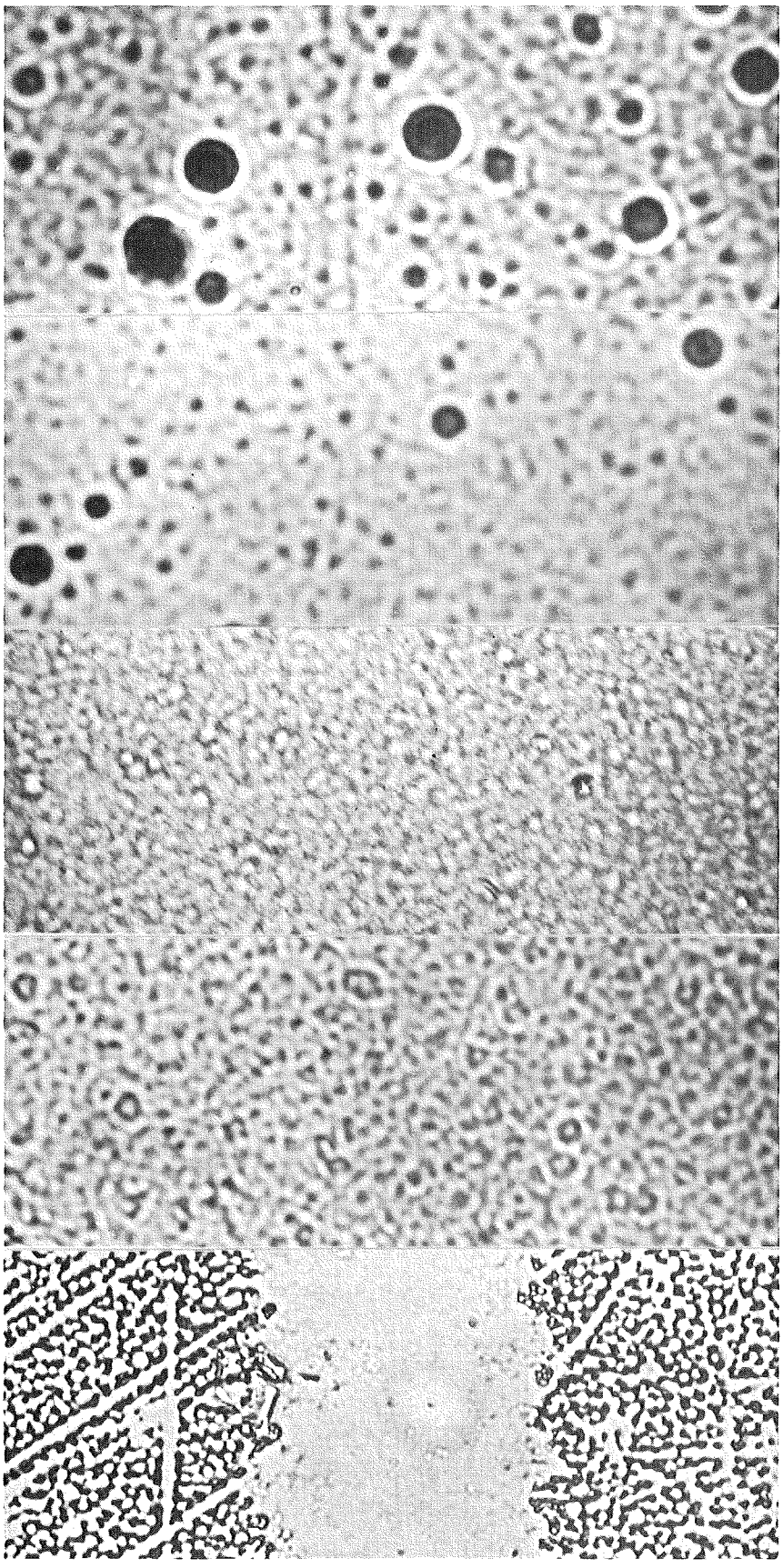

Fig.23: Damage effects of recoils in cellulose acetate after 3 weeks storage time

Figs. 24 and 25 show that at lower temperatures (5°C), the effect of fading is not efficient even at high relative humidities of 97 %. A comparison to Figs. 22 and 23 shows similar fading influences for 50°C or 97 % R.H. Cellulose acetate can therefore be used only under laboratory conditions with a temperature of 25°C and R.H. lower than 53 %.

In Fig. 26, the relative number of tracks was plotted vs. the relative humidity for a storage time of 8 and 21 days and different temperatures. At 25°C, the relative number of tracks slightly decreases if the humidity increases from 0 % to 60 %. Further increase of the relative humidity caused remarkable fading. However at 50°C, high fading was observed when the relative humidity increases from 0 % to 97 %, particularly for long storage time.

Fig. 27 shows the relationship between the relative number of tracks and temperature for cellulose acetate held for 8 and 21 days respectively, at 0 %, 53 % and 97 % relative humidity, respectively. It can be shown in Figs. 27 and 26 that for both storage times, the relative number of tracks is independent of temperature at 0 % relative humidity as well as of humidity at 5°C. At 53 % R.H., the change in the relative number of tracks by increasing temperature is not remarkable if the temperature increased from 5°C to 30°C. If the temperature increased from 30°C to 50°C, the relative number of tracks decreased remarkably. At 97 % relative humidity, a sharp decrease in the relative number of tracks was noticed in the temperature range from 5°C to 50°C. This decrease is much more pronounced for long storage times.

The results presented above show that latent tracks in cellulose acetate are stable at 5°C and 0 % relative humidity for at least 3 weeks after irradiation and before etching. Under laboratory conditions (25°C and 53 % R.H.) only 9 % fading after 3 weeks

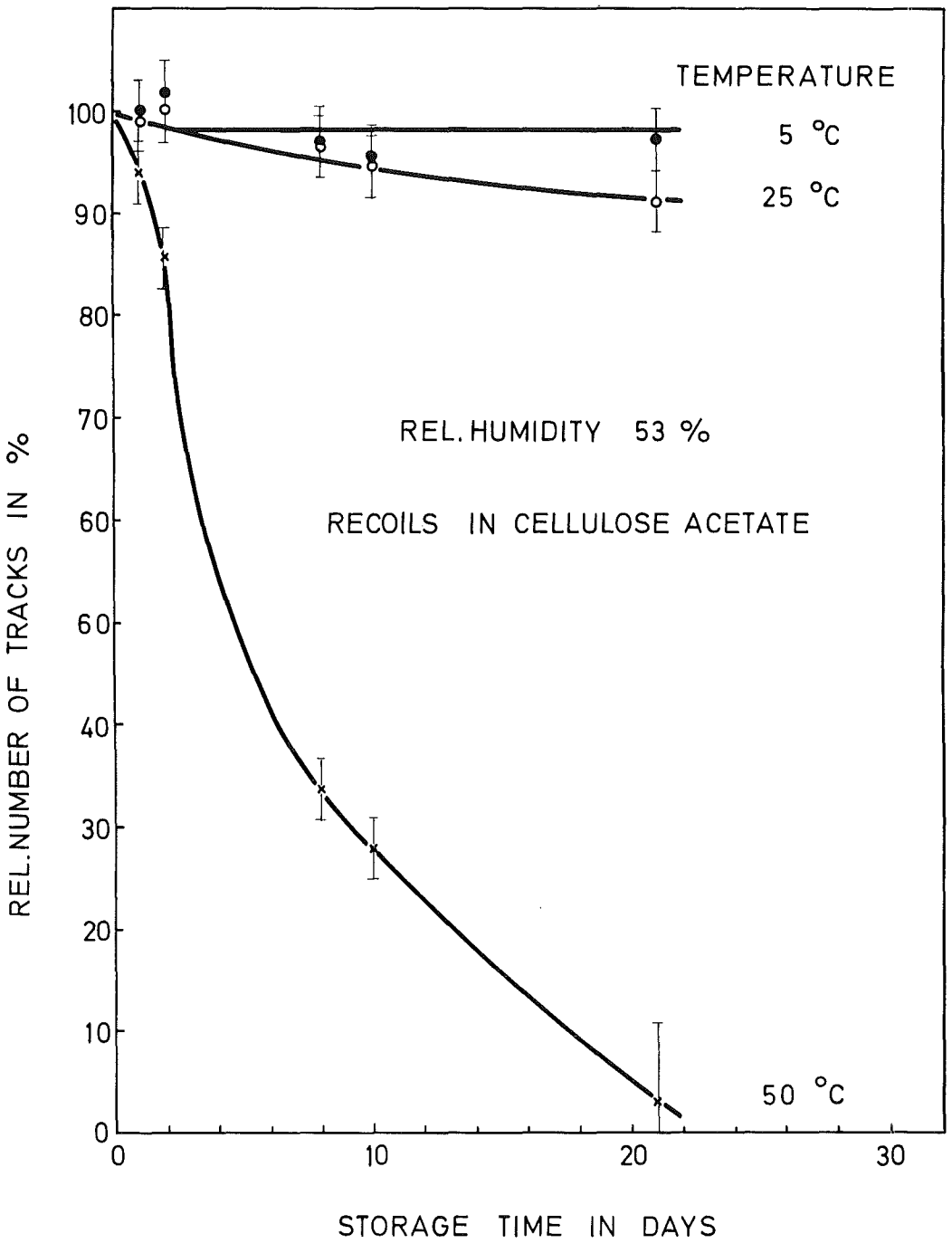

Fig. 24: Fading of recoils in cellulose acetate held at 53 % R.H. and various temperatures

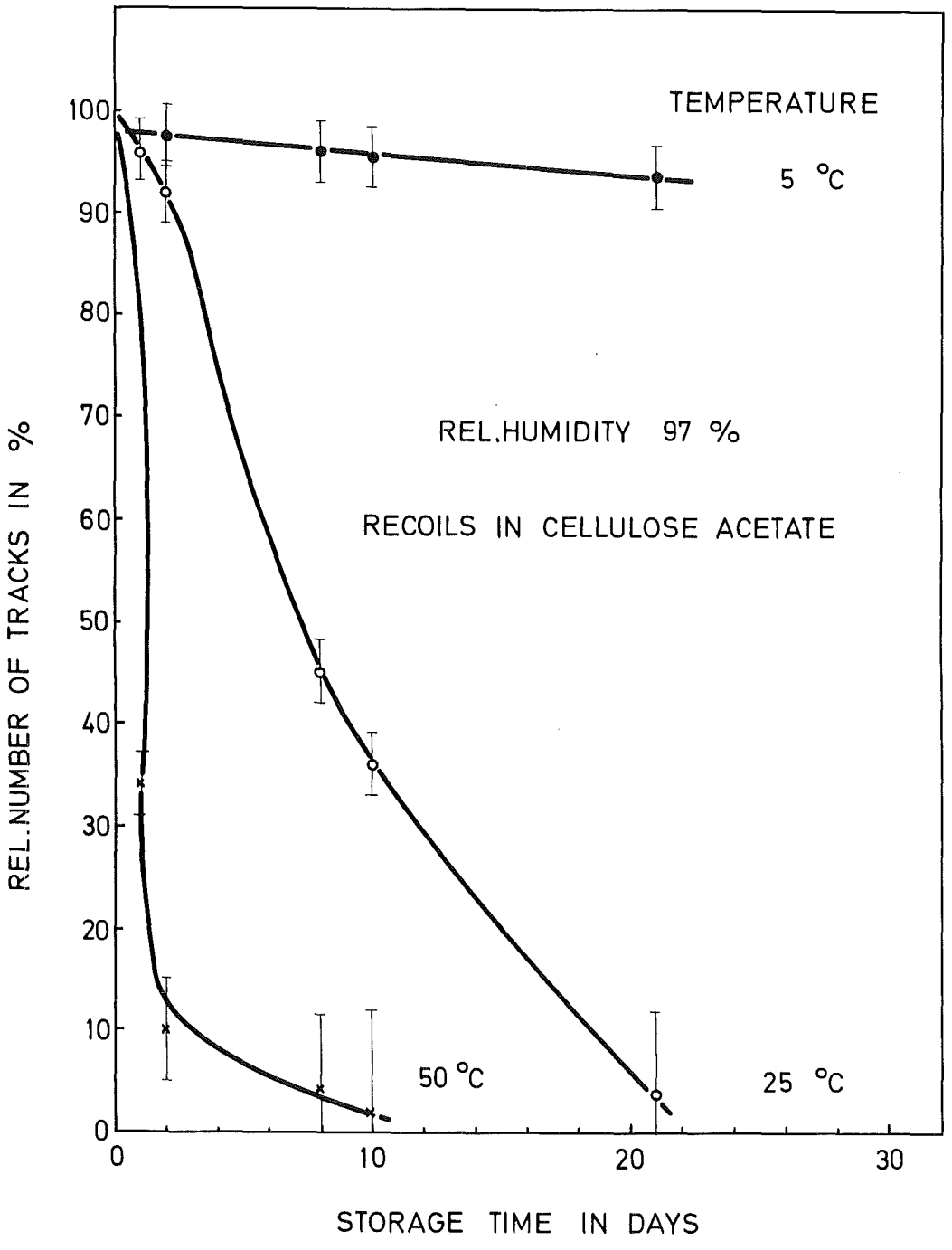

Fig. 25: Fading of recoils in cellulose acetate, held at 97 % R.H. and various temperatures

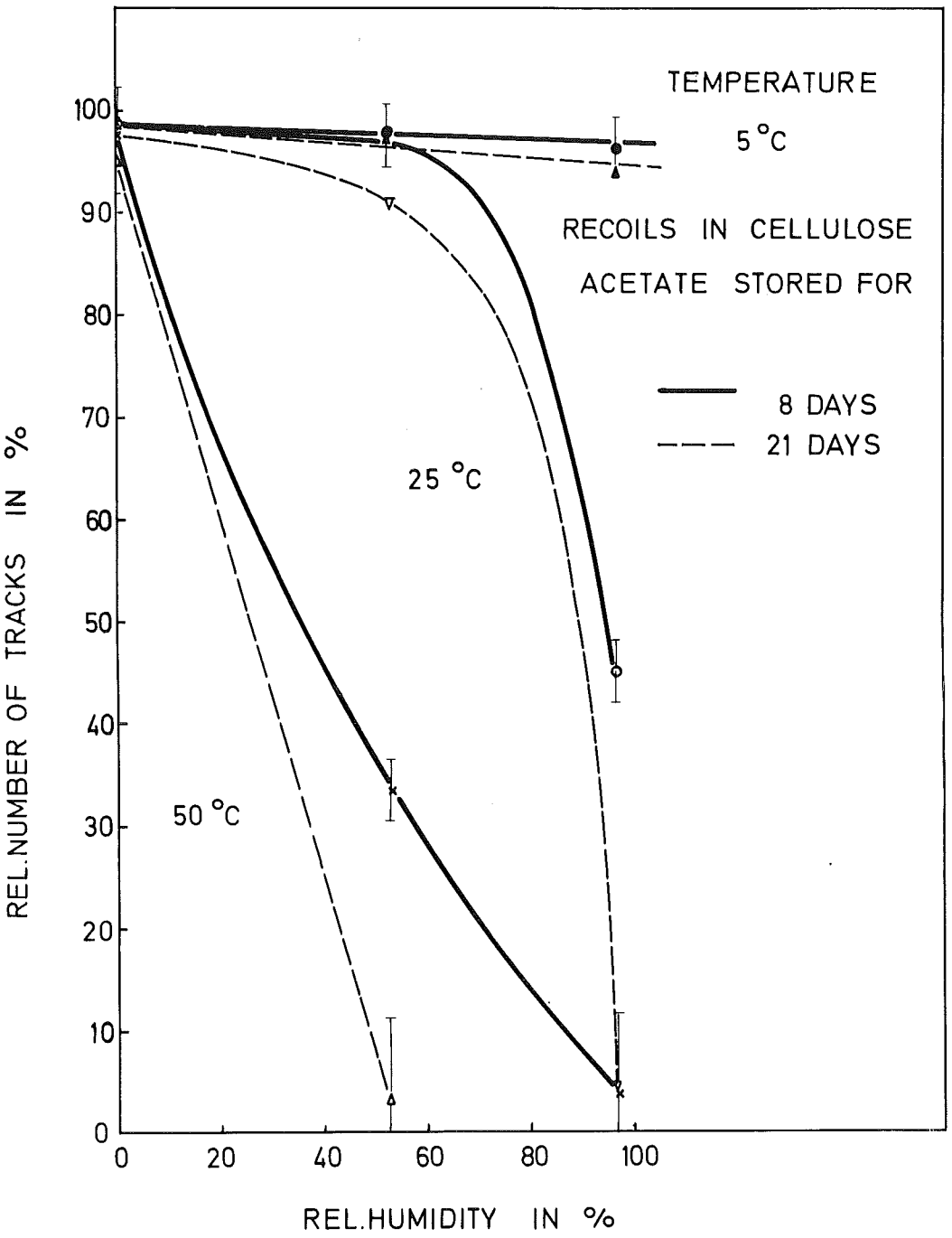

Fig. 26: Relative number of tracks vs. relative humidity for recoils in cellulose acetate stored for 8 and 21 days at different temperatures

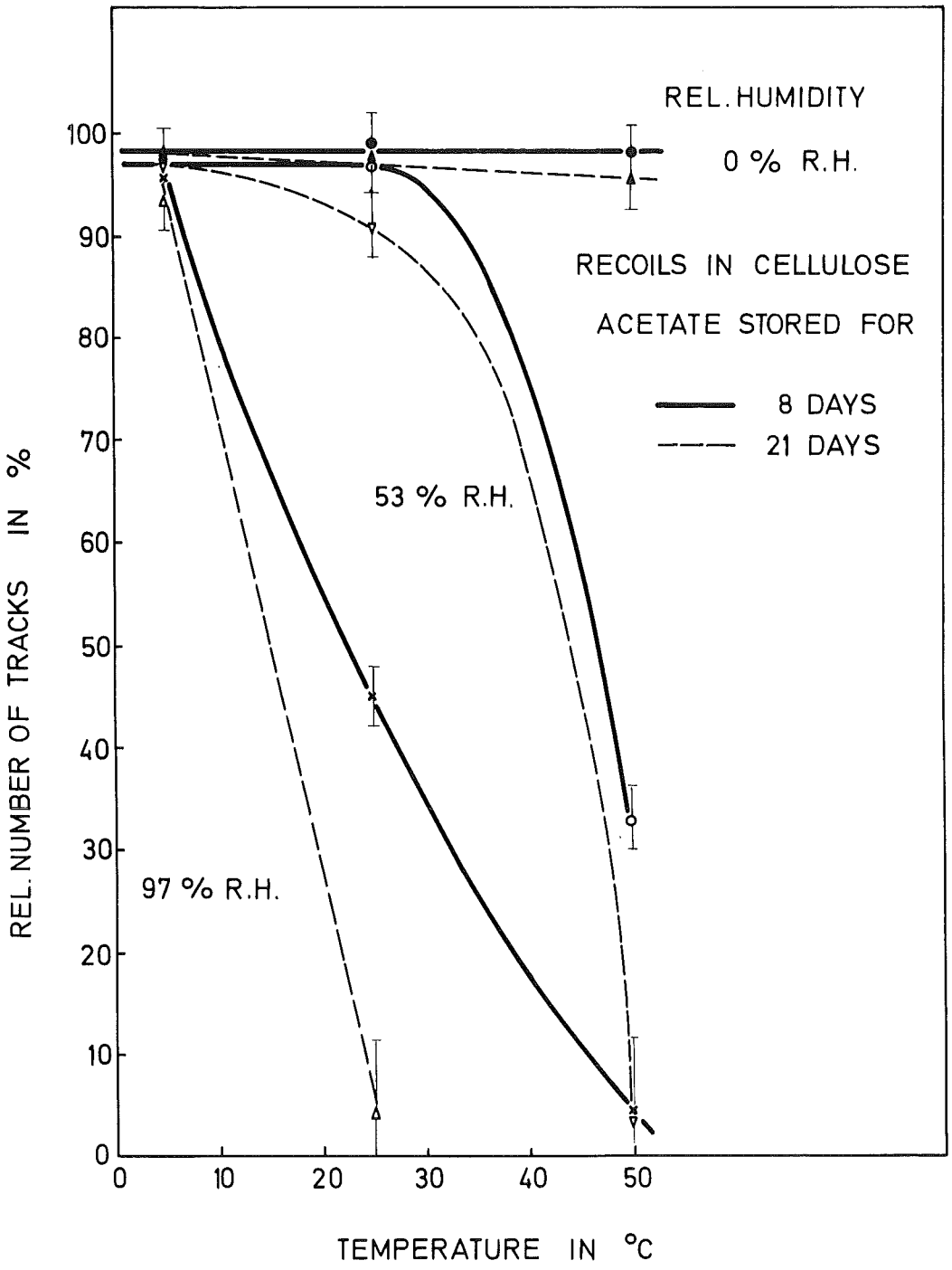

Fig. 27: Relative number of tracks vs. temperature for recoils in cellulose acetate stored for 8 and 21 days at different relative humidities.

was observed. On the other hand, however, high relative humidities may produce high fading even at normal temperature, and 96 % fading was noticed after 3 weeks at 25°C and 97 % R.H. The detector cannot be used at higher temperatures and relative humidities. After 8 days at 50°C and 97 % relative humidity almost all the tracks had disappeared and the beginning of detector damage was found (see also Fig.23).

It was also noticed by Marchetti et al [25] that in some sensitive detectors such as cellulosics, (LR 115 cellulose nitrate made by Kodak Pathé, France), extended storage of the detector at temperatures above 40-50°C may result in permanent changes of their structure and recording properties. After a storage time of 4-15 hours at higher temperatures they found a remarkable fading effect on both track diameter and track density. In cellulose nitrate the latent alpha particle tracks due to the natural background (atmospheric radon) can be annealed by overnight storage at ∿80°C. The influence of atmospherical ozone [31] and of thermal effects on the track registration characteristics - diameter of etch pits, bulk etch rate and thermal track discrimination - in cellulose acetate, cellulose nitrate and polycarbonate was studied by Somogyi [32]. Heinzelmann [33, 34] investigated fading at higher temperatures by variying the etching time for Makrofol E and Triofol.

5.2 In Makrofol E

In Figs 28 and 29 the relative number of tracks were illustrated as a function of storage time for different temperatures at 97 % R.H., and for different relative humidities at 50°C, respectively. Fig. 30 shows the variation of the relative number of tracks with the relative humidity for different temperatures while Fig.31 shows the relative number of tracks as a function of temperature for different relative humidities both for a storage time of 21 days.

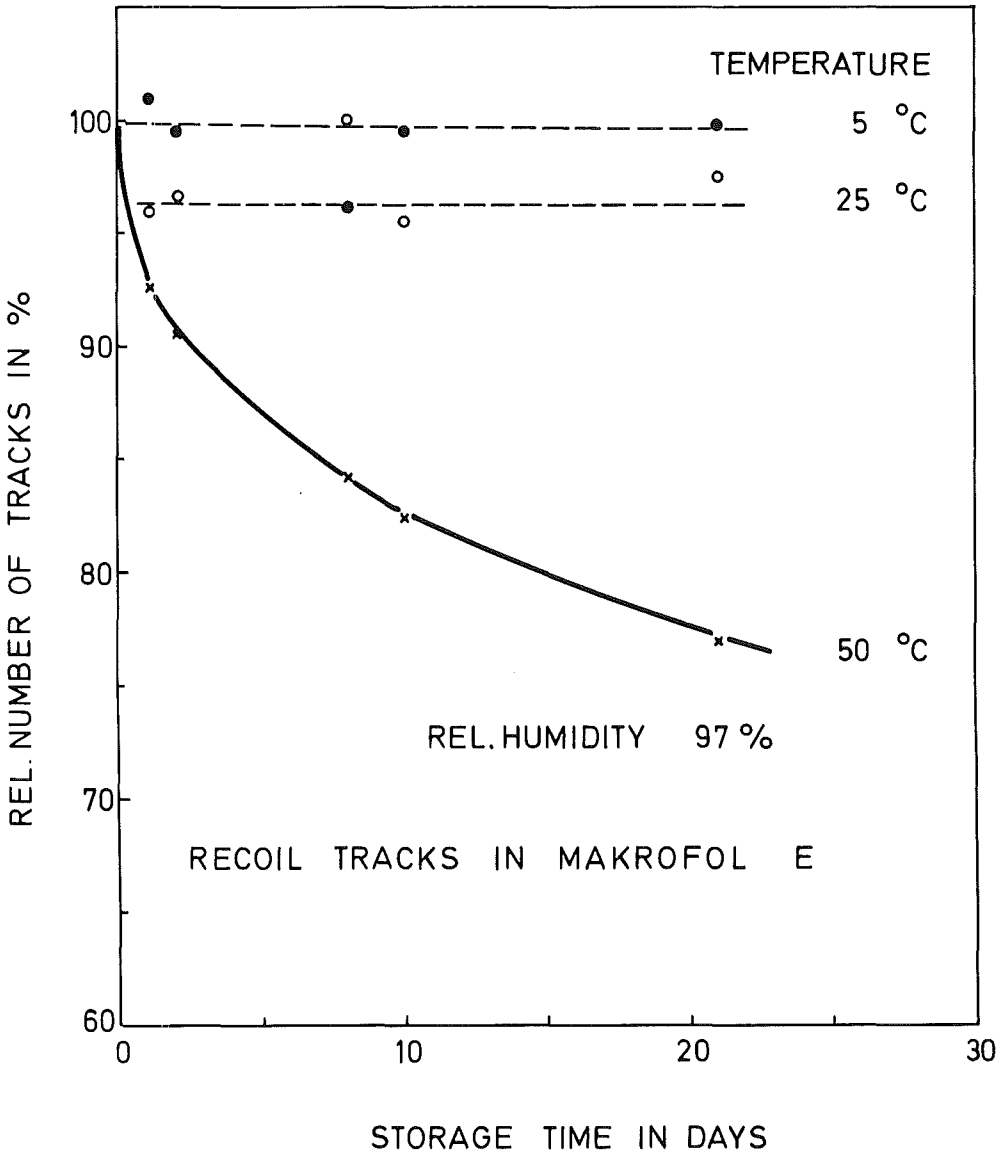

Fig. 28: Variation of the relative number of tracks with storage time for recoils in Makrofol E stored at 97 % R.H. and different temperatures

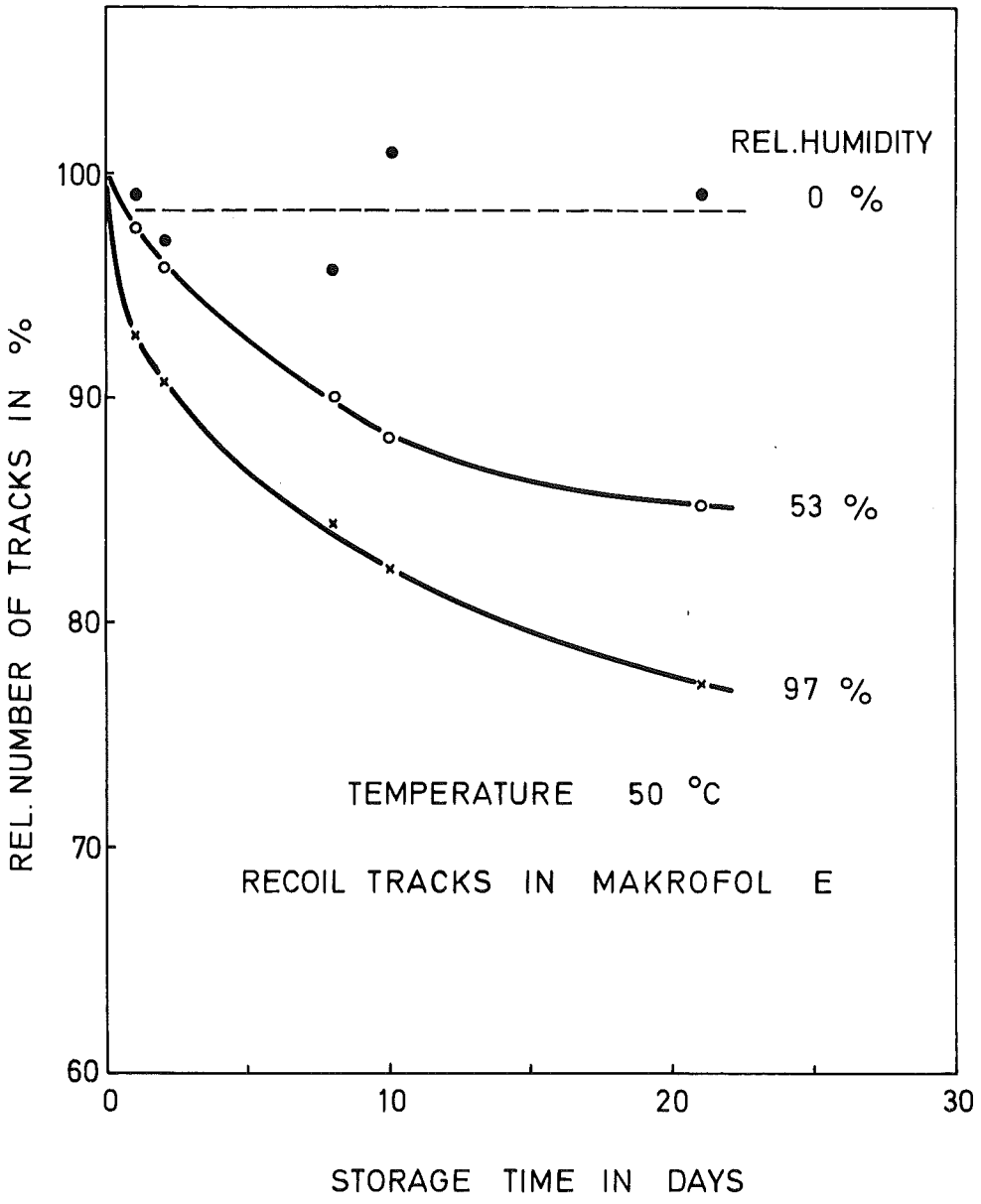

Fig. 29: Variation of the relative number of tracks with storage time for recoils in Makrofol E stored at 50°C and different relative humidities.

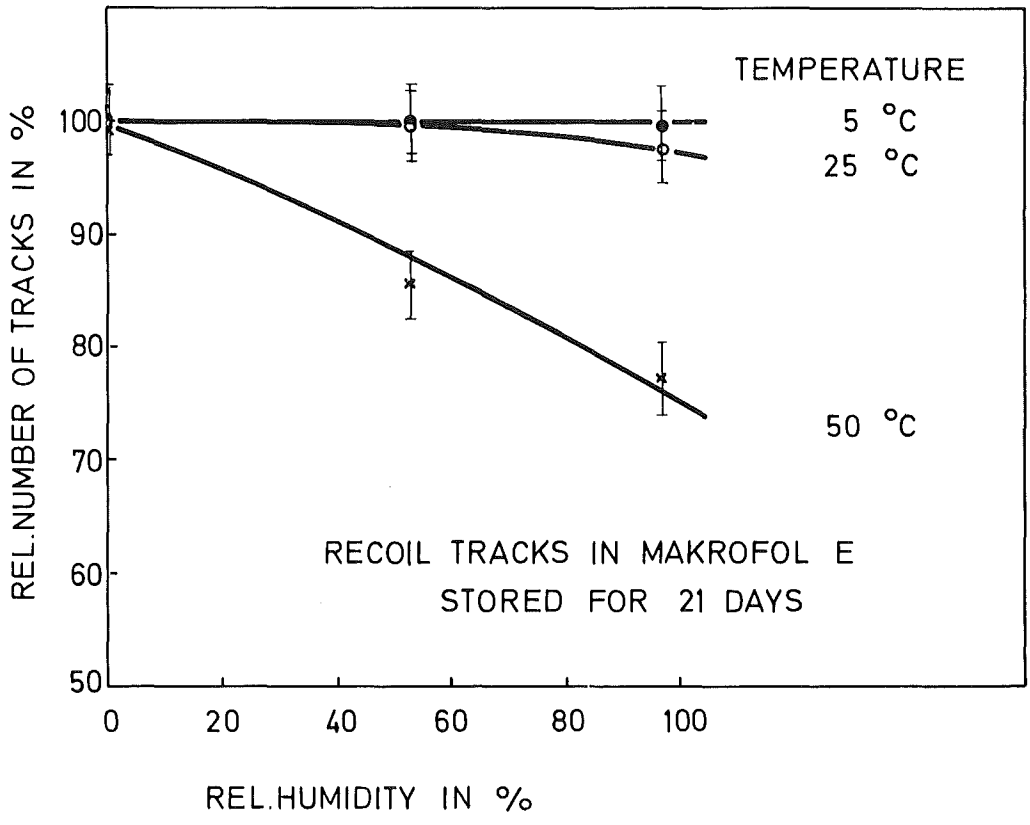

Fig. 30: Relative number of tracks vs. relative humidity for recoils in Makrofol E stored for 21 days at different temperatures

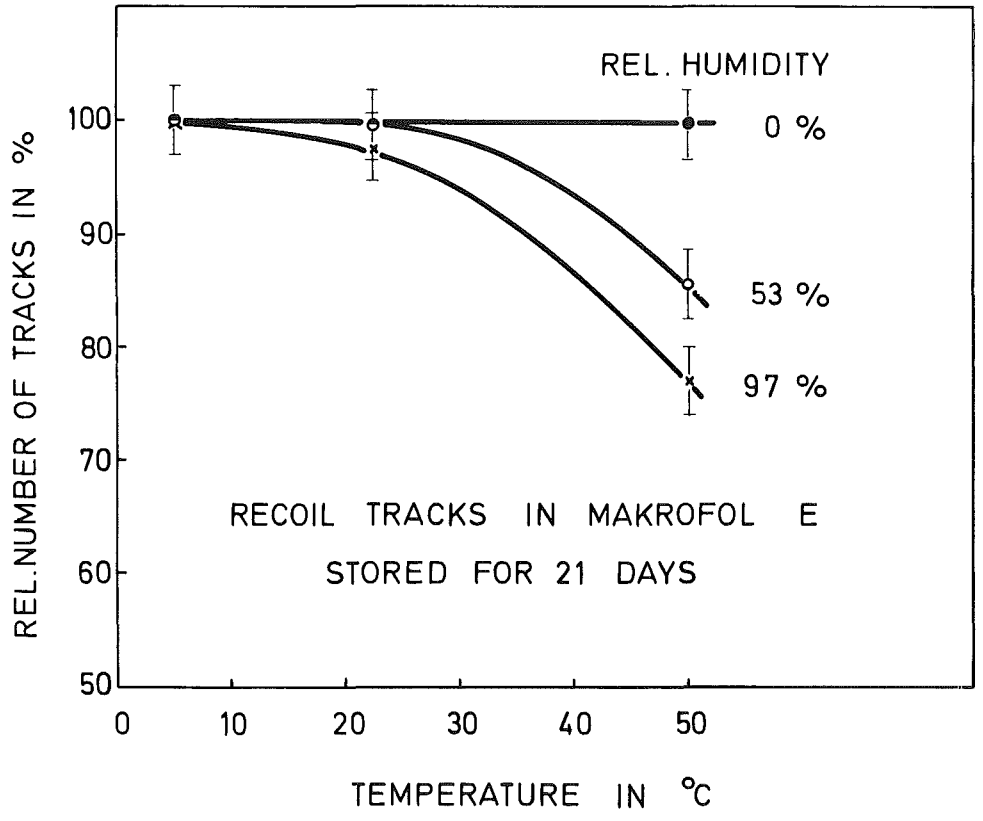

Fig. 31: Relative number of tracks vs. temperature for recoils in Makrofol E stored for 21 days at different relative humidities.

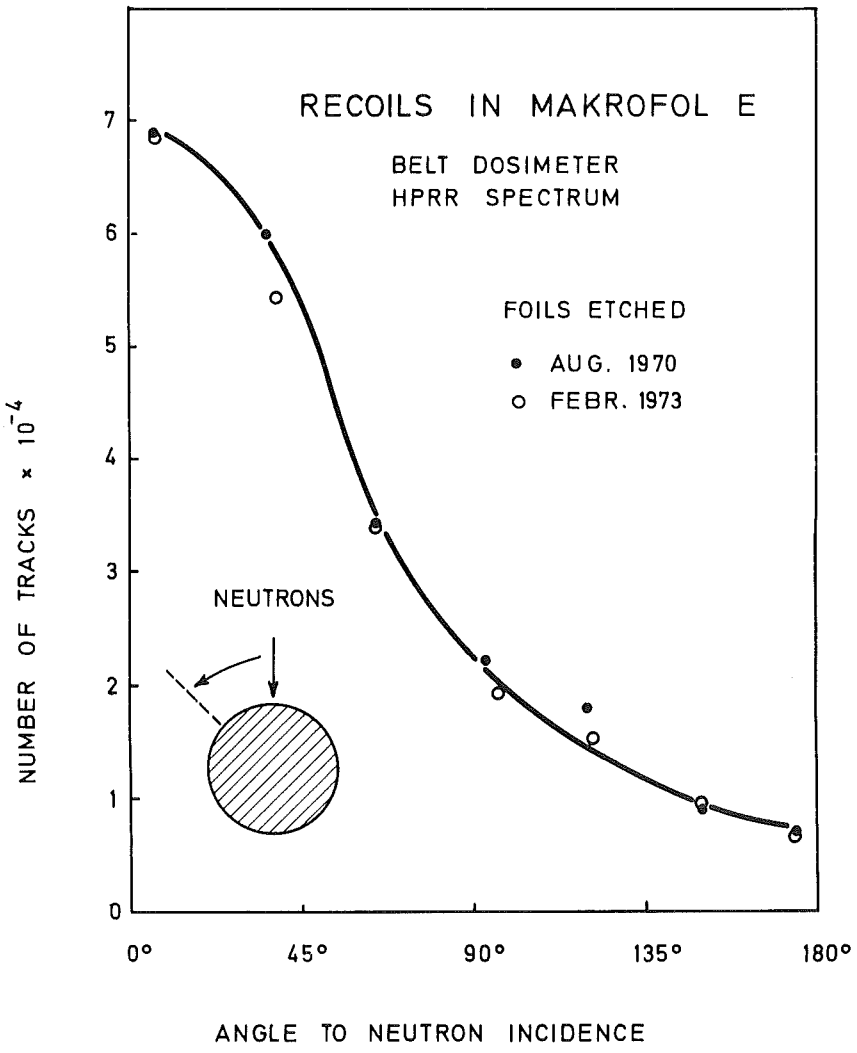

Fig. 32: Number of track as a function of angle to neutron incidence for recoils in Makrofol E

It can be shown from these figures that there is practically no fading at a temperatures of 25°C even for a relative humidity of 97 %. At 25°C and 97 % R.H., the variation of the relative number of tracks after 3 weeks was found to be within the measuring error of 4 %. At 50°C significant fading was noticed, the amount of which depends on both relative humidity and storage time. At 50°C, a loss of 15 % in the relative number of tracks was observed after 3 weeks at 53 % R.H. and a loss of 23 % after 3 weeks at 97 % R.H. respectively.

Studies of long-term fading over a storage time of 2.5 years under laboratory conditions show practically no fading for the latent recoil tracks induced by fission neutrons in Makrofol E. In Fig. 32 the number of recoil tracks are given as a function of angle to the direction of neutron incidence for Makrofol foils which were punched out from a plastic belt after irradiation at the Health Physics Research Reactor, Oak Ridge, USA. Each set of foils were etched in August 1970 and in February 1973, respectively. The mean value for the reduction of track numbers was found to be 7 % over a storage period of 2.5 years. This value includes long-term fading as well as errors from the localisation on the belt and from the etching conditions.

The data presented above showed that recoil tracks in Makrofol E are stable for more than two years under laboratory conditions. The fading is accelerated by higher temperatures above 50°C as well as by high relative humidities of more than 53 %.

6. Conclusions

The investigations of latent fading in nuclear track detectors were performed with the Kodak NTA film, the polycarbonate foil Makrofol KG and Makrofol E and the cellulose acetate foils. In Figs. 33 - 36 a summary of the experimental results is presented for a storage period up to 30 days at a temperature of 25°C and 50°C, and for relative humidities of 53 % and 97 %, respectively.

The NTA film as well as cellulose acetate foils are both detectors with similar fading properties, influenced especially by high air humidity. Makrofol KG and Makrofol E show an excellent long-term stability for fission fragment and neutron induced recoils and α-particles even for high humidities. Significant fading can only be observed at higher temperatures and is slightly affected by (the influence of) humidity.

Based on the results of the fading experiments, the following recommendations can be given for the practical use of these detectors:

- carefully sealed NTA films - which show more than 50 % fading after a period of 3 weeks under laboratory conditions - should only be used for a 2 weeks period in dry cool or moderate climate, but not more than 2 days period in humid and warm climate,
- unsealed, dessicated NTA films can be used for a period of less than 1 week in cool, air-conditioned laboratories with low air humidity,
- cellulose acetate foils - which only show 9 % fading after 3 weeks at 25°C and 53 % R.H., but no tracks at 97 % R.H. - cannot be recommended for the detection of neutron induced recoils and α-particles due to the high influence of humidity and temperature and the corresponding change of the material structure. A short-term use can only be recommended with dessicated foils in additional wrapping at low temperatures and humidities,
- Makrofol E and KG, a track etching detector with long-term stability - which shows no significant fading below 50°C, only 23 % fading after a 3 weeks period at 50°C and 97 % R.H. - can be used even in an extremely hot and humid climate. Under laboratory conditions the storage time can be extended over a period of more than one year.

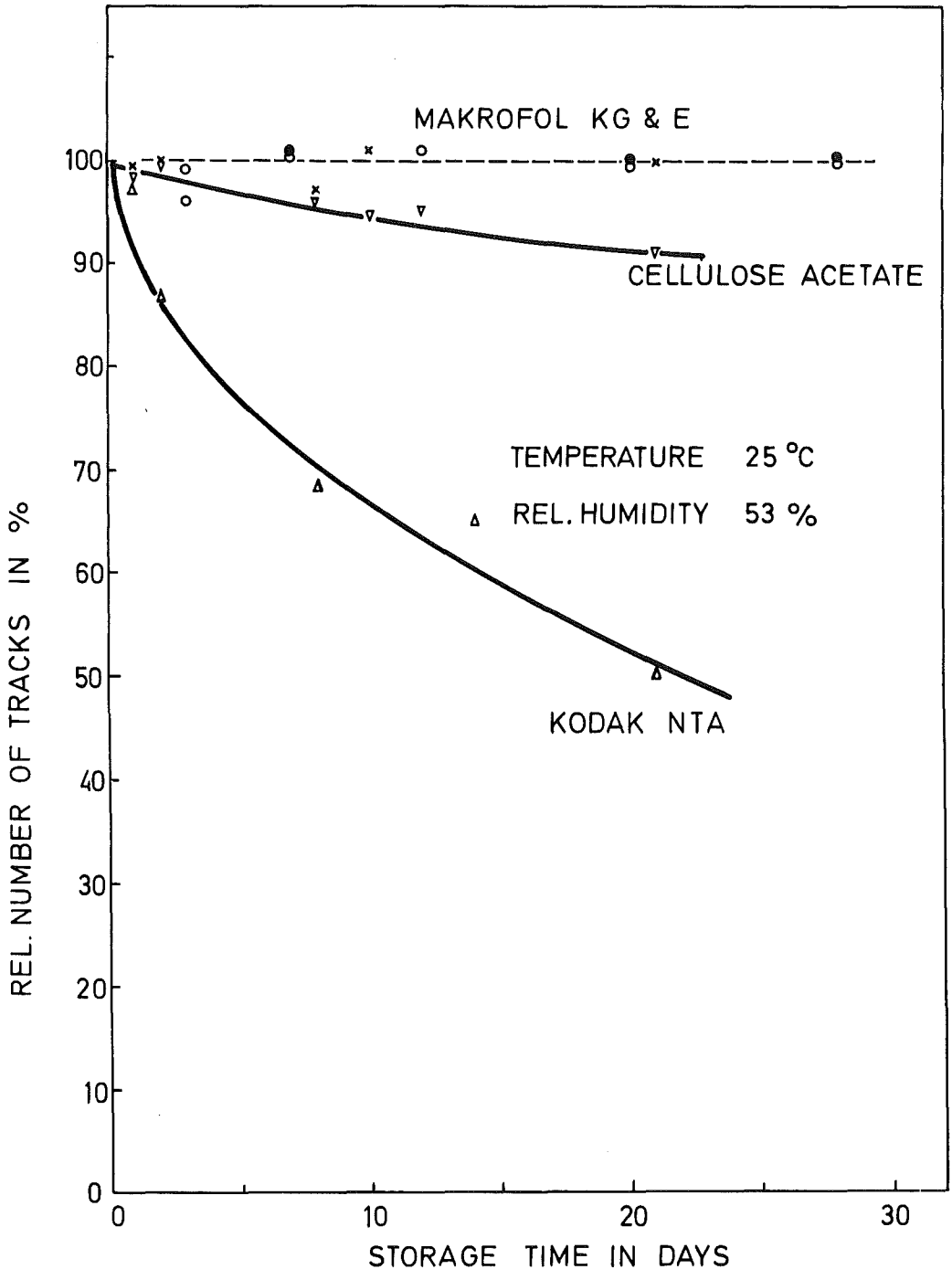

Fig. 33: Comparison of latent fading in Kodak NTA, Makrofol and cellulose acetate, held at 25°C and 53 % R.H.

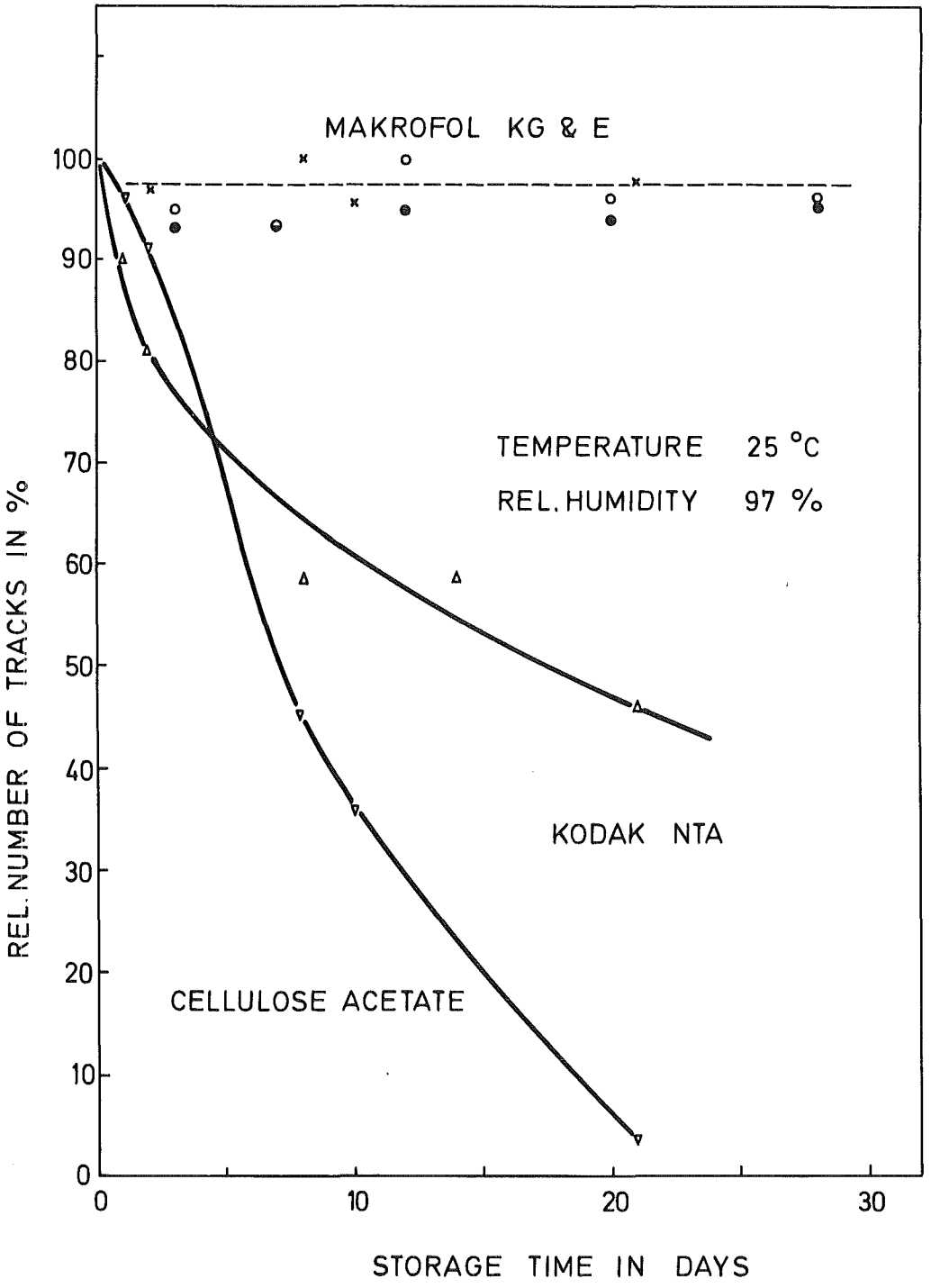

Fig. 34: Latent fading in Kodak NTA, Makrofol and cellulose acetate, stored at 25°C and 97 % R.H.

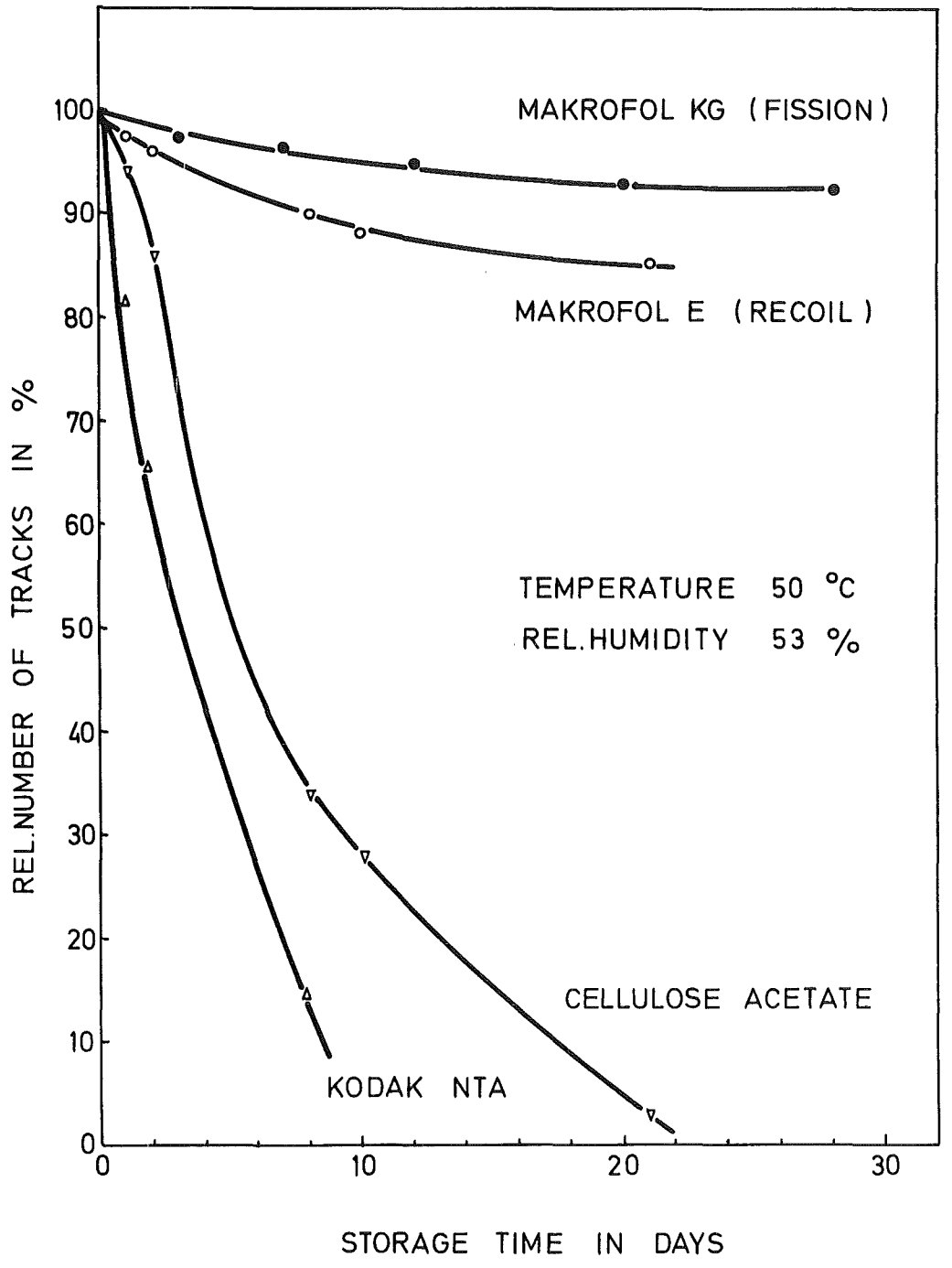

Fig. 35: Relative number of tracks as a function of storage time for Kodak NTA, Makrofol and cellulose acetate, stored at 50°C and 53 % R.H.

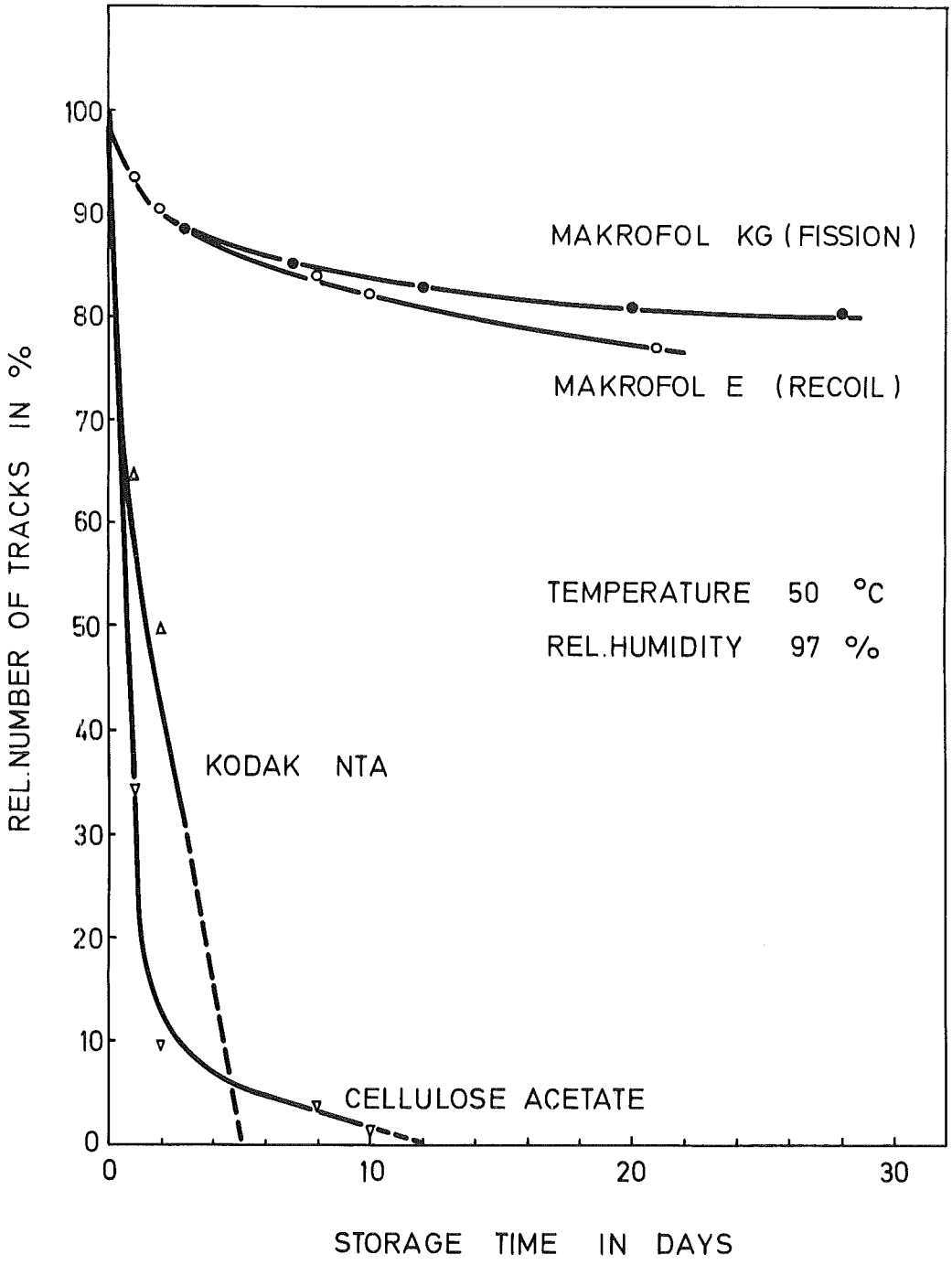

Fig. 36: Relative number of track vs. storage time for Kodak NTA, Makrofol and cellulose acetate, stored at 50°C and 97 % R.H.

From the practical point of view, the NTA film should be used in an additional sealing of an aluminium-plastic foil. The film should not be sealed when equilibrated under laboratory conditions. To achieve equilibrium with the environmental conditions, the film in the original factory wrapping should be stored for a period of 12 days. Dessicated films must be carefully sealed in a dry, cool atmosphere. Otherwise, fading of the sealed film is higher than that of an unsealed film due to the humidity content inside the sealing. A special treatment or detector wrapping is not necessary for track etching detectors except for cellulose acetate. The fading results found with control films which were exposed and stored under actual environmental conditions, can practically be used as a correction factorsfor a given storage period, if the fading influence in routine monitoring does not exceed 50 %. The need for higher accuracy and lower fading correction in the order of 10-20 % is one of the most important aspects when selecting an optimum detector.

Track etching detectors are coming more in use, above all for special applications, for instance in neutron spectroscopy using a threshold detector combination. Based on an inquiry, Griffith [26] reported that in 1972 more than 118 laboratories we using track etching methods. However, the difficult and time consuming in handling of these detector type prevents an extended use of track etching detectors in routine monitoring. The results of recent investigations [27-29] show that the albedo neutron dosimeter using LiF detectors will become an alternative for the measurement of fast neutrons even with more sufficient dosimetric properties, for instance lower energy dependence without energy threshold, extended dose range, simple reading and insignificant fading.

The investigations of latent fading, the results of which are presented in this report, were only performed with film and track etching detectors excluding other solid-state detectors such as phosphate glass dosimeters or thermoluminescence dosimeters, which nevertheless have been in routine use for more than 8 years. But the long-term stability of RPL and TL dosimeters will not be affected by influences of humidity to such an extent as for the detectors discussed in this paper.

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References

[1] J.S. Cheka: Nucleonics, 12, 40 (1954)

[2] J.P. Cusimano: Latent Image Fading in Personnal Neutron Monitoring Film at NRTS, IDO-12031, 1963

[3] S. Hagsgard, C.O. Widell: Personal Neutron Monitoring at AB Atomenergie: Neutron Dosimetry, Vol. II, 249-258, IAEA, Vienna, 1963

[4] R. Mc Carthy, V. Mejdhal: Personnel Dosimetry Practice at Riso: Risö Report No. 78, 1964

[5] H. Dresel: Bestimmung der Belastung durch schnelle Neutronen mit Kernspurfilmen im Rahmen der routinemässigen Personenüberwachung, Kerntechnik 11, 498-500 (1961)

[6] G. Portal: Dosimetrie des neutrons rapides au moyen d'emulsions a protons de recul, Personnel Dosimetry Techniques for External Radiation, 219-235, Symposium, Madrid, 1963

[7] P. Amadesi, A. Cavallini, O. Rimondi: Taratura del film badge per neutroni veloci, Minerva Nucleare 5, N12, 323-336 (1961)

[8] C.A. Turner: Nucleonics, 24, 62-63 (1966)

[9] J.B. Kahle, E.N. Arnett, C.T. Meyer: Latent Image Fading in Personnel Monitoring Neutron Film, Health Physics, 17, 735-736 (1969)

[10] R.L. Fleischer, P.B. Price, R.M. Walker: Science 149, 383 (1965)

[11] A. Cavallini, G. Busuoli: Proc. OECD/ENEA Symp. Pers. Radiat. Protection, Stockholm 293 (1967)

[12] J. Jasiak, T. Musialowicz: Nukleonika 17, 647, 1972

[13] E. Piesch: Atompraxis 9, 179, 1963

[14] R.L. Lehmann: UCRL-9513, 1961

[15] E. Piesch: Atompraxis 10,
Direct Information 4/64, 1964

[16] Jósefowicz: Proc. IAEA Symp. Neutron Monitoring, 1973

[17] E. Piesch: Proc. IAEA Symp. Advances Phys. Det., 399, 1971

[18] A. Wexler, S. Hasegawa: J. Res. NBS-53, 19, 1954

[19] E. Piesch: Proc. Int. Conf. Congress Clermont Ferrand III-66, 1969, Zeiss Informationen 18, Nr. 76, 58, 1970

[20] K. Becker: Report ORNL-TM-4297, 1973

[21] P.N. Krishnamoorthy, et.al.: Proc. IAEA-Symp. Neutron Monitoring, 1973

[22] J. Jasiak, T. Musialowicz: Proc. IAEA-Symp. Neutron Monitoring 1973

[23] C.H. Distenfeld, J.R. Klemish: Report BNL 17452, 1972

[24] Seguin: Proc. IAEA Symp. Neutron Monitoring 1973

[25] Maschetti et al: Proc. 8th Int. Conf. Nucl. Photo and Track Etching, Bucharest, 1972

[26] Griffith, R.V.: UCRL-51362, 1973

[27] J.E. Hoy: Health Physics 23, 385, 1972

[28] E. Piesch, B. Burgkhardt: Proc. IAEA Symp. Neutron Monitoring, 1973

[29] E. Piesch, B.Burgkhardt: Proc. IRPA Congress Washington, 1973

[30] R.E. Zelac: Health Physics, 15, 545, 1968

[31] G. Somogyi: Radiation Effects 16, 233, 1972

[32] G. Somogyi: Radiation Effects 16, 245, 1972

[33] M. Heinzelmann: Report JÜL-603-ST, p.191-194, 1969

[34] M. Heinzelmann: Proc. 1st Symp. Neutron Dosimetry in Biol. and Med., EUR 4896 d-f-e Vol.1 p.315, 1972