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LONG-TERM STABILITY OF FILM, TLD, AND  
OTHER INTEGRATING DOSIMETERS IN  
WARM AND HUMID CLIMATES

Klaus Becker

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HEALTH PHYSICS DIVISION

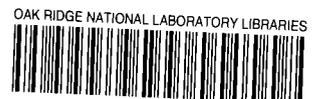
LONG-TERM STABILITY OF FILM, TLD, AND OTHER INTEGRATING  
DOSIMETERS IN WARM AND HUMID CLIMATES

Klaus Becker

AUGUST 1973

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Long-Term Stability of Film, TLD, and Other Integrating  
Dosimeters in Warm and Humid Climates\*)\*\*)

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ABSTRACT

With the increasing use of radiation sources in hot and/or humid climates, the long-term stability of dosimeters selected for personnel, environmental, and postal intercomparison dosimetry is becoming a matter of world-wide concern. Laboratory tests with various film, TLD, RPL (glass), silicon diode, TSEE and track etching dosimeters at ORNL and other laboratories are reviewed and compared with the results of field tests, in particular a recent study involving the storage of different gamma monitoring films with or without additional protective sealing, LiF:Mg,Ti (TLD-100), and CaSO<sub>4</sub>:Dy at seven locations in four Latin American countries. The observed fading rates range from 0 to 100 % for the Kodak Type 2 and 3 films, and from 0 to 23 % for the TLD phosphors, depending on the climate of the location. In four of the seven locations, unsealed films should not be used in X- and gamma-radiation personnel dosimetry (in two of those places, even optimum sealing did not prevent 54-89 % fading). Extremes in storage temperature are most critical for TLD, extremes in relative humidity for the film fading. The results and the consequences to be drawn are discussed. Apparently, even carefully sealed dosimeter films cannot be used in the climate prevailing at many locations. Some desirable follow-up studies are outlined.

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\*) Research sponsored by the U.S. Atomic Energy Commission under contract with the Union Carbide Corporation.

\*\* )Dedicated to Boris Rajewsky on occasion of his 80th birthday.

## Introduction

There are at least four areas of increasing practical concern in which the long-term stability of sensitive, integrating radiation detectors under severe climatic conditions is of crucial importance:

1. Personnel radiation monitoring programs in regions which have, at least during part of the year, hot and humid (tropical) weather, as it is the case in most Third World Countries, and in the Southern parts of Europe, the United States, and the Soviet Union.
2. Environmental monitoring for establishing population exposure levels in areas with an increased natural or artificial background in the above regions;
3. Intercomparison programs in which photon, electron and neutron radiation sources used in cancer therapy, industrial processing, etc., are calibrated by mailing detectors over large distances for standard irradiation, with evaluation of the detectors in a central laboratory; and,
4. to a somewhat lesser degree, *in-vivo* dosimetry in man or warm-blooded animals with implanted or *in-utero* dosimeters.

In each case, many technical, economical, organizational, and sometimes even "political", psychological and educational factors have been considered in the choice of optimum detector systems. But there should be no doubt that the detector(s) finally selected have to meet certain minimum performance criteria. The accuracy and reliability requirements vary for different applications. While a  $\pm 30\%$  overall accuracy of dose measurements is usually sufficient for personnel dosimetry, accuracies

of better than  $\pm 5\%$  are required for calibration studies (for a more detailed recent discussion of these factors, see ref. 1,2).

So far, large-scale radiation monitoring programs have been mostly restricted to the highly developed countries with a moderate climate and air-conditioned hospitals, laboratories, etc., whose temperatures and humidities rarely deviate much from the "normal", comfortable values of 18-24°C and 40-75 % relative humidity. This situation is, however, changing. With the increasing worldwide concern about radiation protection and the increasing use of radiation sources in medicine, industry and research, the number and volume of personnel monitoring programs, as well as of area monitoring, and the need for intercalibrations of radiation sources in poorly equipped installations is presently undergoing a rapid expansion. Consequently, the results of most of the numerous fading tests which have been carried out previously, insuring (with a few exceptions such as the NTA film and some TLD phosphors) that the stability of the detectors is sufficient for monitoring periods of up to one or a few months in a moderate climate, are not relevant for more severe climates.

Actual field conditions are different from constant laboratory conditions mostly in the extremes in temperature and humidity which are frequently encountered during part of the total storage period, even if the average temperatures and humidities are close to "normal". In Santa Fé, Argentina, for example (a location involved in our tests in Latin America), the average temperature in February 1973 was 17.8°C, but peak temperatures reached 38.2°C. One has to keep in mind

- a. that fading accelerates rapidly with increasing temperature and humidity (a few hours at 40°C may be equivalent to one month at 20°C in the total induced TL fading, and a few hours at 100 % relative humidity can completely anneal an NTA film); and
- b. that there are apparently temperature and humidity thresholds for certain types of damage to the detectors (only little thermal fogging has been observed in dosimeter films below 45-50°C).

Even in a short-term intercomparison study by mail, involving, for example, several hours of exposure of the detectors to high temperatures in the mail compartment of a plane standing in the noon sun at a tropical airport, the fading produced during this period can easily outweigh the effects of weeks of storage in an air-conditioned room. Furthermore, conditions which fluctuate on a daily and seasonal basis may have a non-additive effect in some detectors. For instance, the full penetration of humidity through film wrappings (or the release of humidity from the film) is known to proceed with a delay time which may amount to several days. Obviously, even careful laboratory tests cannot simulate field conditions completely, and estimates based solely on such tests may be rather inaccurate. It is the purpose of this report to analyze the available laboratory data, compare them with the results of field tests, and to discuss some of the consequences for radiation protection programs as well as future research needs.

## 2. Photographic Film

Photographic emulsion and its radiation response characteristics have been studied in great detail (for compilations, see ref. 1,3).

Extended storage of fresh films in a humid and warm environment is known to lead to

- a. fogging (increase of the background optical density, O.D., mostly caused by thermally induced formation of developable silver centers, or thermally accelerated chemical fogging);
- b. irreversible changes in sensitivity and other response characteristics (4);
- c. "sticking" of the emulsion layer to the film wrapping, with partial or total destruction of the emulsion occurring when the pack is opened in the darkroom (Fig. 1); and
- d. microbiological growth in gelatin (a good culture medium for various common bacteria and fungi), which may also result in total destruction of the sensitive layer.

It is a fairly common observation that dosimeter films which had been left near hot radiators, in the glove compartment of cars parked in the sun in summer, etc., exhibit a high O.D. which can be mistaken for a gamma-radiation exposure. The mechanism of thermal fogging, and the role played by humidity is not yet completely understood, but it appears that

- a. little thermal fogging occurs at temperatures below 45 to 50°C for short storage periods, while above this temperature range it rapidly accelerates (Fig. 2); and
- b. maximum thermal fogging takes place at relative humidities around 50 %, probably because latent-image regression (fading) compensates for the spurious latent-image formation at higher humidities (5).



Fig. 1 Developed Kodak Personal Monitoring Film Type 2, heavily damaged during several weeks of storage at 30°C and 95 % relative humidity.

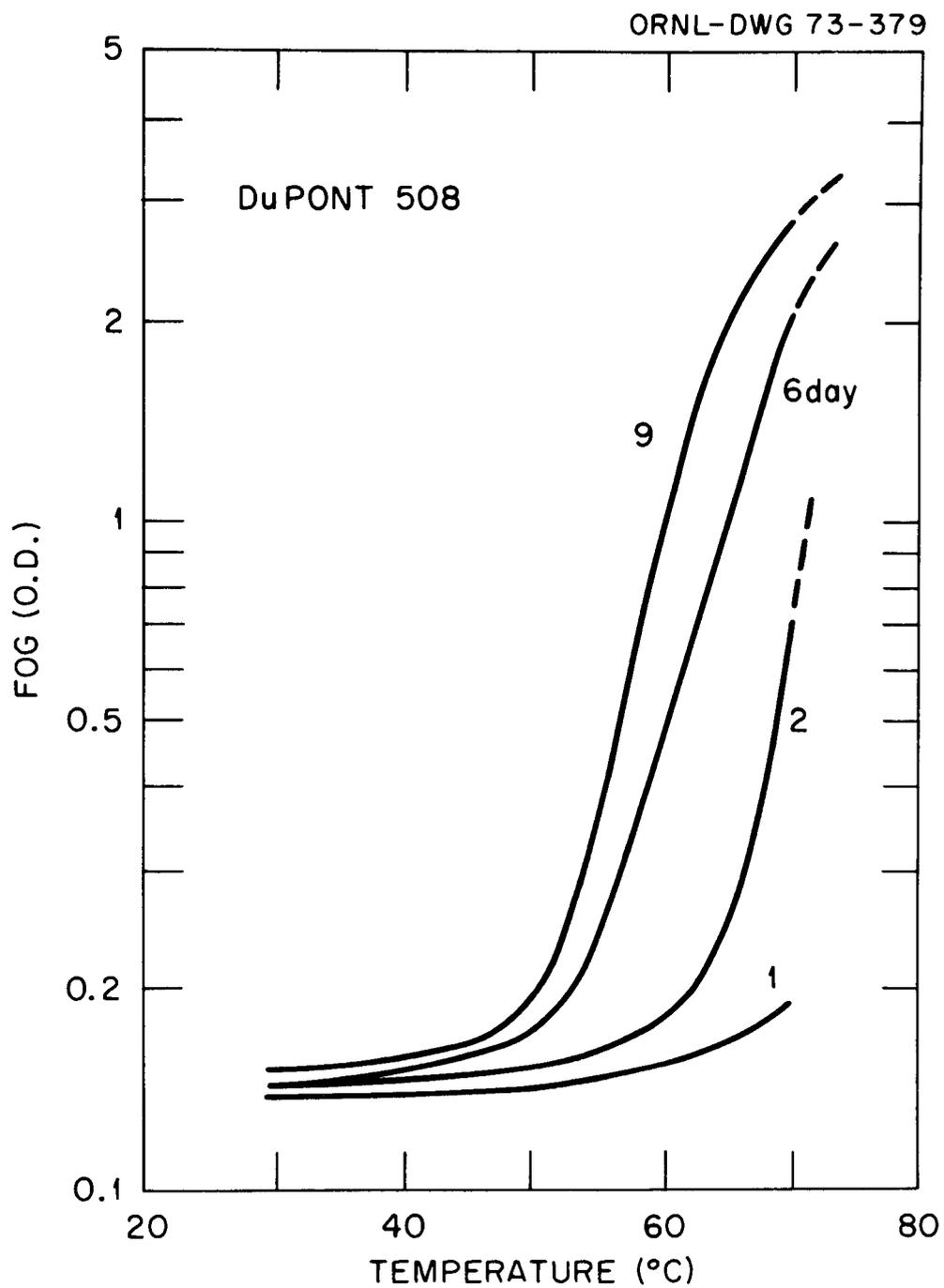


Fig. 2 Thermal fogging of an unirradiated dosimeter film during storage for various times at different elevated temperatures (after 5).

Much more serious than these effects is the fading which occurs after exposure. Fading results partly from thermal dissociation of the latent development centers (the centers consist of aggregates of  $\sim 4-10$  silver atoms in the silver halide crystal), but mostly (about 90 % of the total fading) because of the combined chemical action of oxygen and humidity on these latent-image specks. One can visualize the process as a silver-catalyzed reaction  $2 \text{H}_2\text{O} + \text{O}_2 \rightarrow 2 \text{H}_2\text{O}_2$ , in which the resulting  $\text{H}_2\text{O}_2$  destroys the catalyst, namely the silver development centers. They are usually located at the microcrystal (grain) surface even if the original ionization took place in the grain interior. Fine-grained emulsions with their larger surface area frequently exhibit more fading than coarse-grained ones, but there are exceptions to this rule.

Being a chemical process, fading is strongly accelerated by increases in the storage temperature. The kinetics are not simple, but the fading of the radiation-induced O.D. can usually be approximated as the log of time. The superimposed effects of fading and fogging may lead to an "equilibrium" O.D. in dosimeter films which is fairly independent of their actual radiation exposure. This effect, as observed in the insensitive emulsion of a Kodak Type 2 film pack during storage at  $30^\circ\text{C}$  and 95 % relative humidity (the less stable sensitive emulsion was totally destroyed after this period) is illustrated in Fig. 3.

Fading has been known since 1910, and many publications have been devoted to this subject, of which only a few can be quoted here<sup>\*)</sup>.

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\*) In addition to the published information, several film dosimetry services are known to have carried out laboratory and/or field stability tests, which did in some cases lead them to conclude that "fading is no problem". As no details on these unpublished studies are known, they cannot be discussed here.

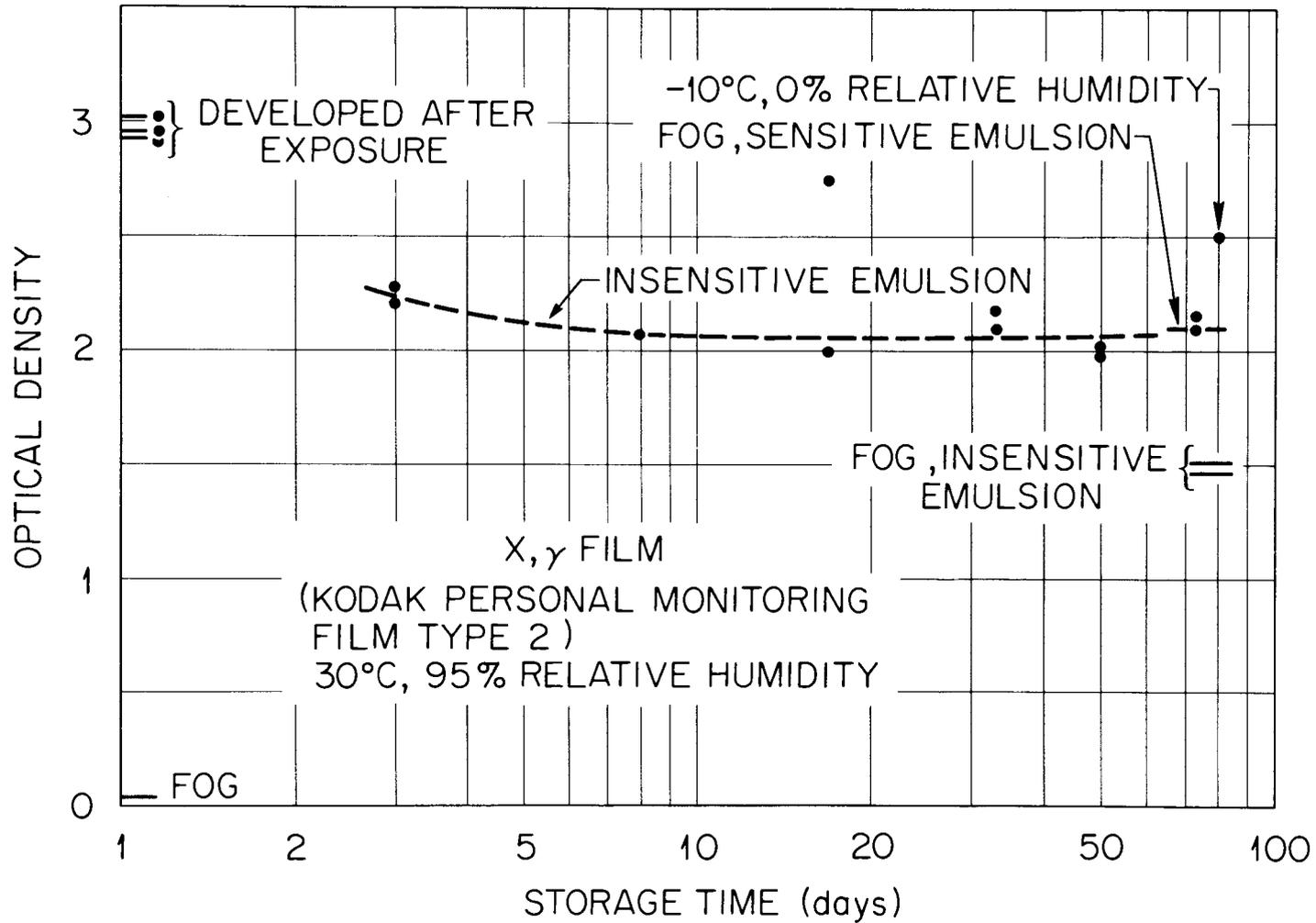


Fig. 3 Fading and fogging in the insensitive emulsion of a Kodak Personal Monitoring Film Type 2 during three months of storage in a simulated tropical climate at 30°C, 95 % relative humidity.

Several reports indicate that only relatively little fading takes place under "normal" conditions in coarse-grained, highly sensitive X-ray films which are commonly used as sensitive X- and  $\gamma$ -ray detectors in personnel dosimetry. In most of these reports, the storage climate is not well specified, but it may be assumed that most of the tests have been carried out in a fairly dry, comfortable laboratory climate. At high relative humidities, however, substantial fading has also been observed in such films in laboratory as well as in field tests, for example in the Kodak RM film (Fig. 4) and in the Kodak Personnel Monitoring Type 2 film (Fig. 5). In another study, during 10 days at 25°C, or during 6 days at 35°C (Fig. 6), 100 % fading in moist air has been found. In the latter study, the apparent gamma dose reading actually became negative after about one week because the background fog was also reduced by fading.

Besides its dependence on humidity and temperature, the fading rate also depends on the dose level and on photon energy. In one experiment involving a slow X-ray film, for example, no fading was observed during one month after a low-energy X-ray exposure, but 30 % fading occurred after  $^{60}\text{Co}$  gamma radiation exposure (7). Most published data on dosimeter film stability, in particular older ones, are not very reliable and certainly not comparable partly because of differences in the detailed experimental conditions, but also because emulsions and film packaging are frequently modified by the manufacturers.

Fading is much more pronounced in the fine-grained nuclear track emulsions which are still widely used in fast-neutron personnel dosimetry. The effect is well-documented, but as the experimental conditions have

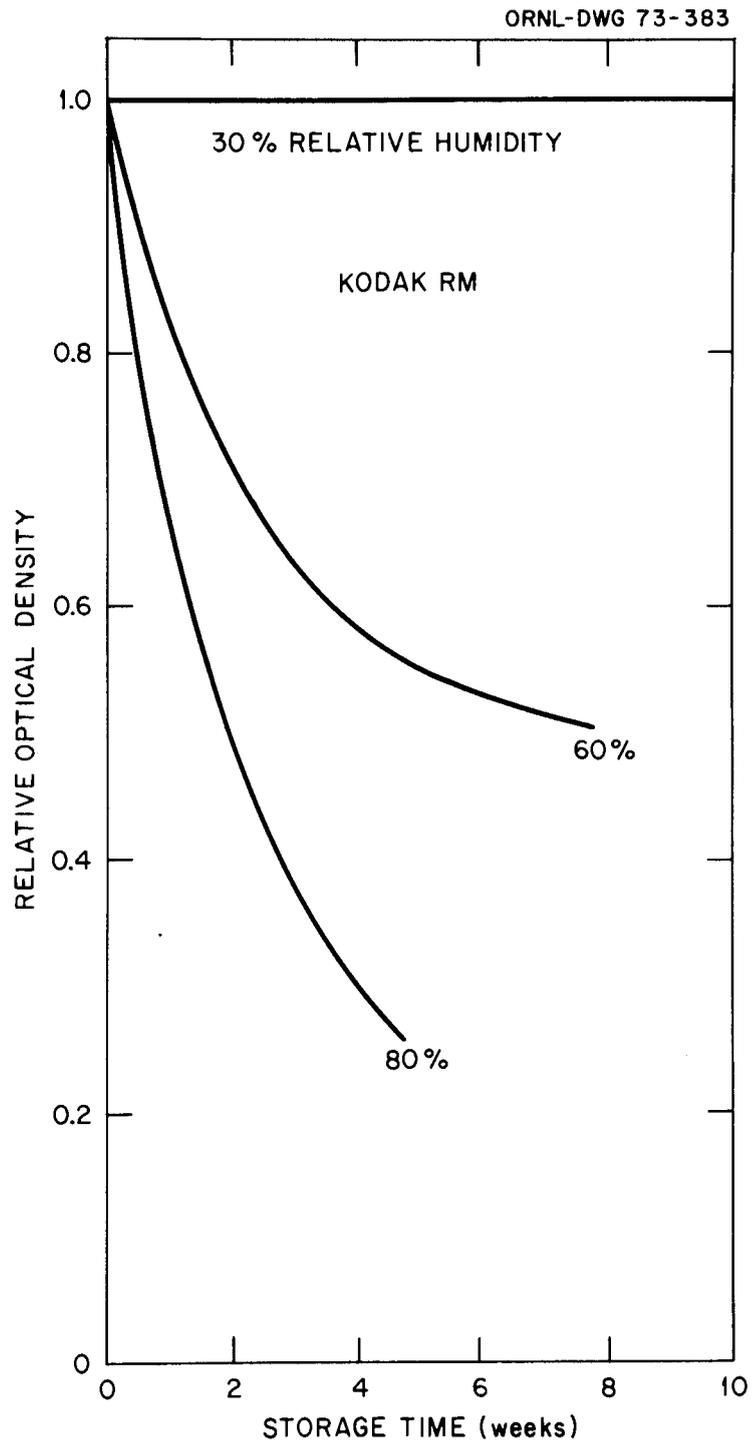


Fig. 4 Latent image fading in a Kodak RM dosimeter film at 30°C and different relative humidities (after 32).

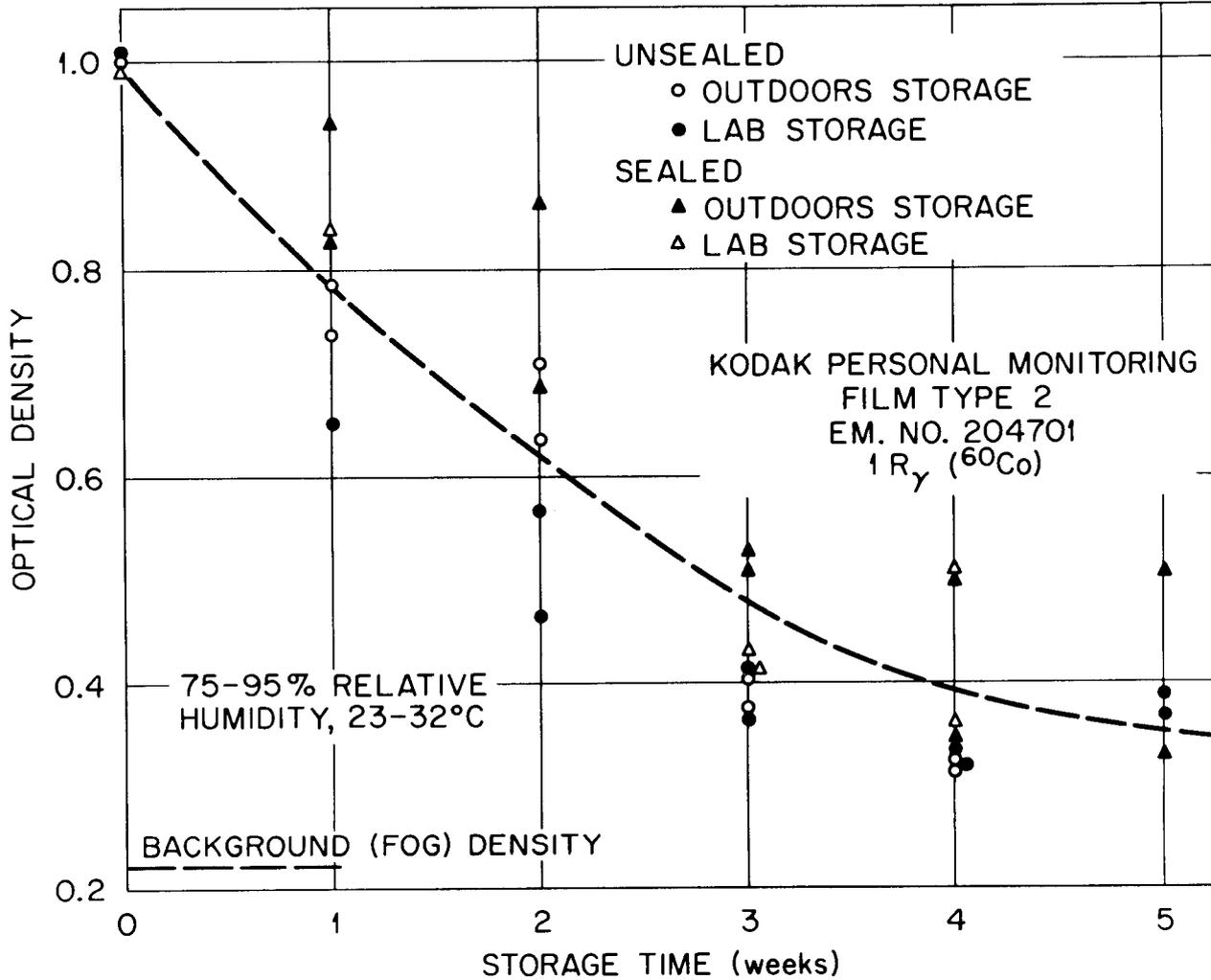


Fig. 5 Optical density of a gamma-irradiated Kodak Personal Monitoring Film Type 2 (sensitive emulsion) as a function of storage time between exposure and (simultaneous) processing of the films; storage in standard laboratory atmosphere, and in a protected space outdoors with or without additional sealing in a thin polyethylene bag, in a subtropical climate. Note the wide scattering of the O.D.s in the stored films.

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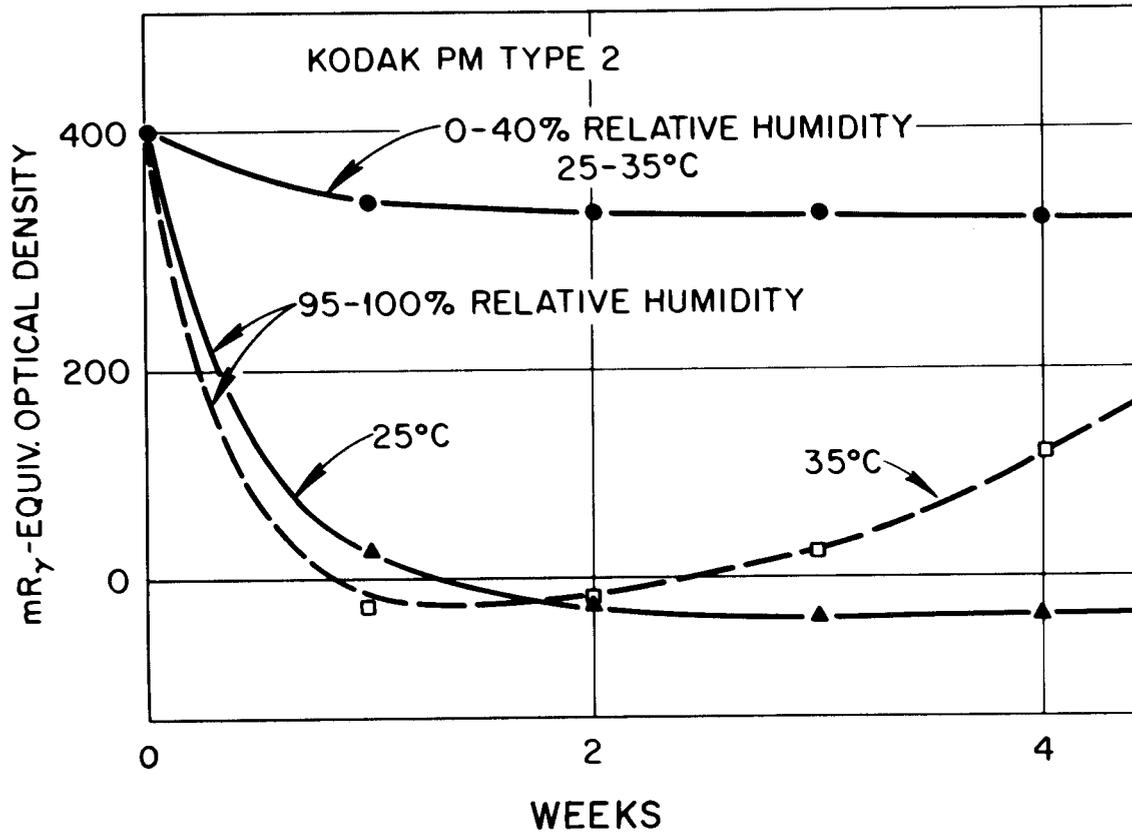


Fig. 6 Fading of the total optical density in a Kodak PM Type 2 film after exposure to 400 mR of gamma radiation during storage at different climates (after 6).

been often ill-defined, or are for other reasons (such as the use of different types of emulsions) not comparable, there has been some disagreement in the reported fading rates and the effect of protective measures (see, for example, ref. 8-29, 91). One will also obtain quite different results for the fading of the O.D. on the one hand, and the disappearance of visible tracks on the other hand, because a long, dense track may be still visible after some of its grains have disappeared. The visibility of tracks also strongly depends on their length (e.g. on the effective neutron energy), and the fog density (neutron to gamma dose ratio).

Furthermore, the films may not have been equilibrated with their environment at the beginning of a fading test: As can be seen in Fig. 7, the visible track density in a film in equilibrium with an average laboratory climate (20°C, 75 % relative humidity) decreases in 10 days by a factor of two. Equilibrium is the more realistic condition, because a personnel dosimeter is usually worn for some time prior to the neutron exposure. But if one starts with a dry film, it may require a few days before the humidity fully penetrates the wrapping (at Brookhaven National Laboratory, for example, ~ 60 % fading has been observed during two weeks at 71 % relative humidity and 32°C in pre-desiccated films; at 22-26°C and 43 to 61 %, it amounted to about 50 % during one month - 91). In Fig. 8, the track fading of equilibrated films is given for a constant temperature (22°C) for different relative humidities. These data are in good agreement with other recent measurements (for example, ref. 25).

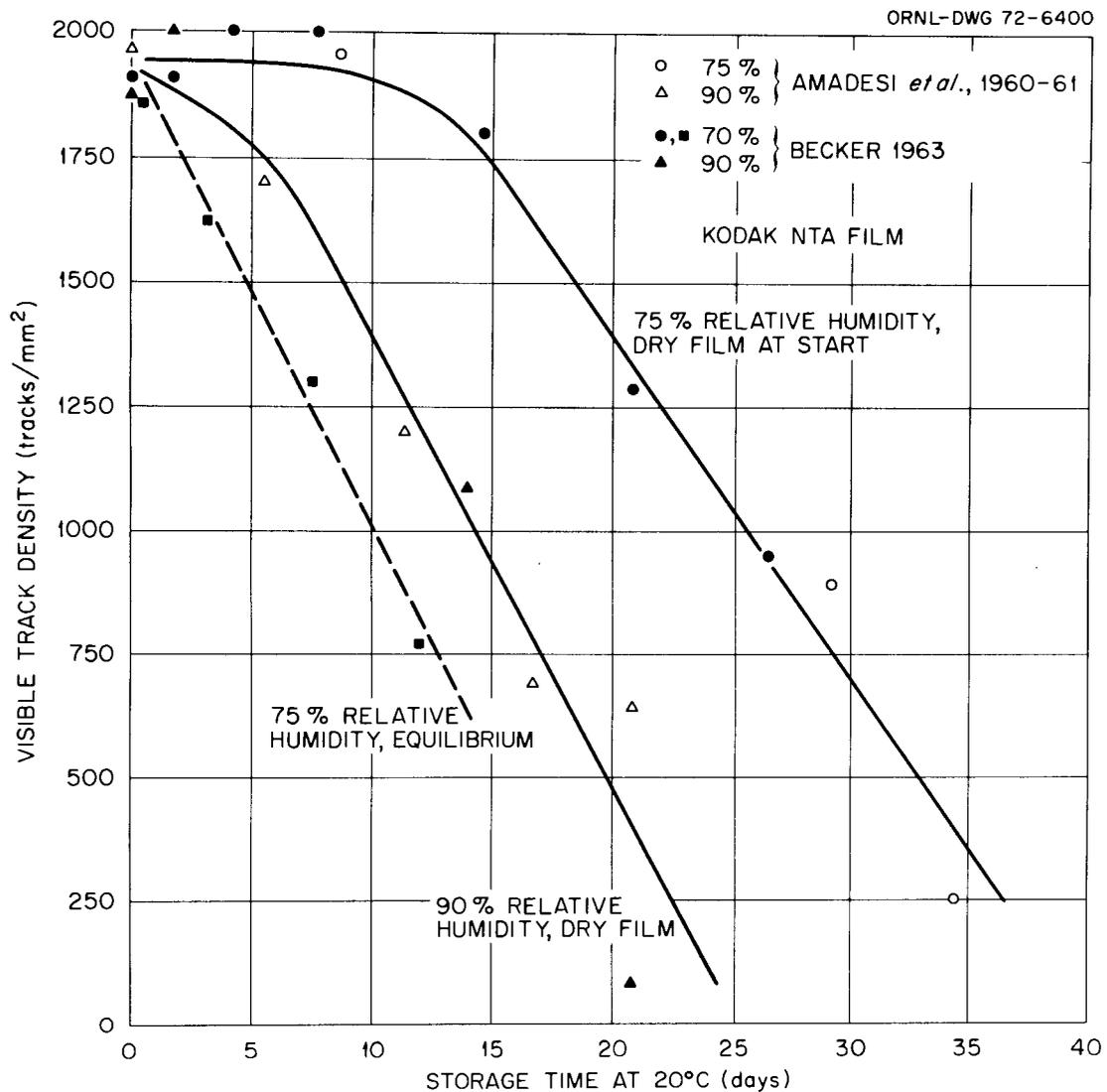


Fig. 7 Visible recoil proton track density in Kodak NTA film as a function of storage time at 20°C, and 75 % and 90 % relative humidity, if the film (without additional wrapping) is irradiated after careful drying; and at 75 % relative humidity, if permitted to equilibrate prior to neutron irradiation.

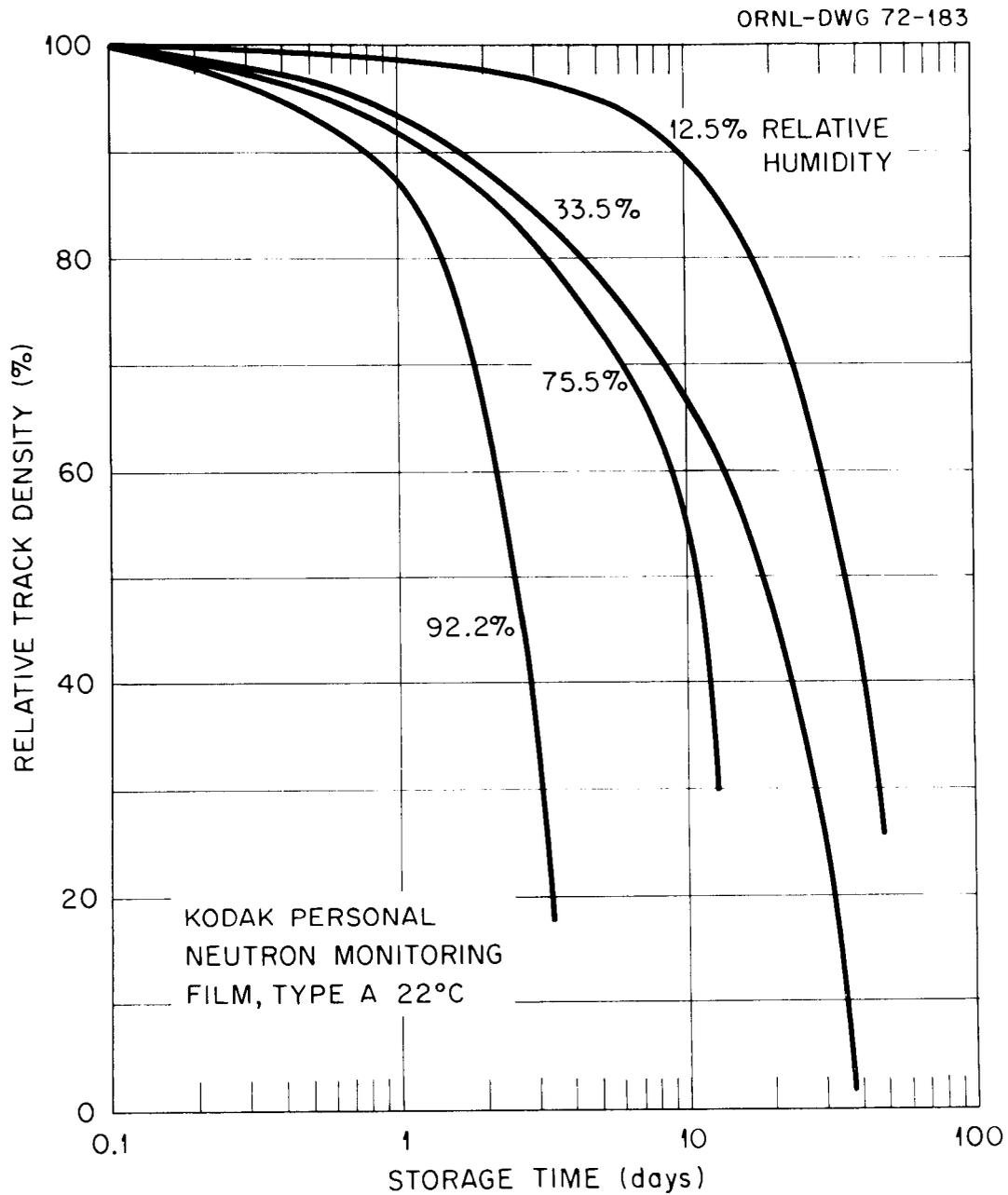


Fig. 8 Relative visible recoil proton track density in Kodak NTA film exposed to Po/Be neutrons as a function of storage time at different humidities.

The accelerating effect of increased temperature on fading is illustrated in Fig. 9. As can be seen, at 35°C (which is not an uncommon temperature in subtropical and tropical countries), 50 % track fading occurs in one week even at the very low humidity of 38 %, instead of the ~ 82 % relative humidity necessary to induce this fading rate at 22°C. Field tests confirm these laboratory results: in a subtropical climate, 50 % fading was observed in equilibrated track films without additional wrapping within ~ 1.5 days, and ~ 90 % of the radiation effect had disappeared after one week (Fig. 10).

It is, in principle, possible to reduce fading somewhat by modifications of the emulsion, or suppression of surface development centers (the internal image is more stable). More effective has been the exclusion of oxygen, or, even better, of humidity. Unfortunately, gelatin is a rather hygroscopic substance which absorbs large quantities of water (at 75 % humidity, 20 % by weight - Fig. 11).

Many investigators have tried to exclude humidity by sealing dry films into polymer foils. The permeability of polymers to water vapor varies widely, and strongly depends on various factors such as their density, crystallinity, orientation and crosslinking of the molecules, and additives. As can be seen in Table 1, ethyl cellulose is more permeable than certain types of Teflon by a factor of more than  $10^4$ . Many polymers are obviously rather "transparent" to water vapor, but even the most "impermeable" foil has a measurable permeability (for additional information, see ref. 92). The rate of water vapor transmission depends on temperature and on the vapor pressure gradient across the foil. The driving force is largest with a completely dry film inside, and a high

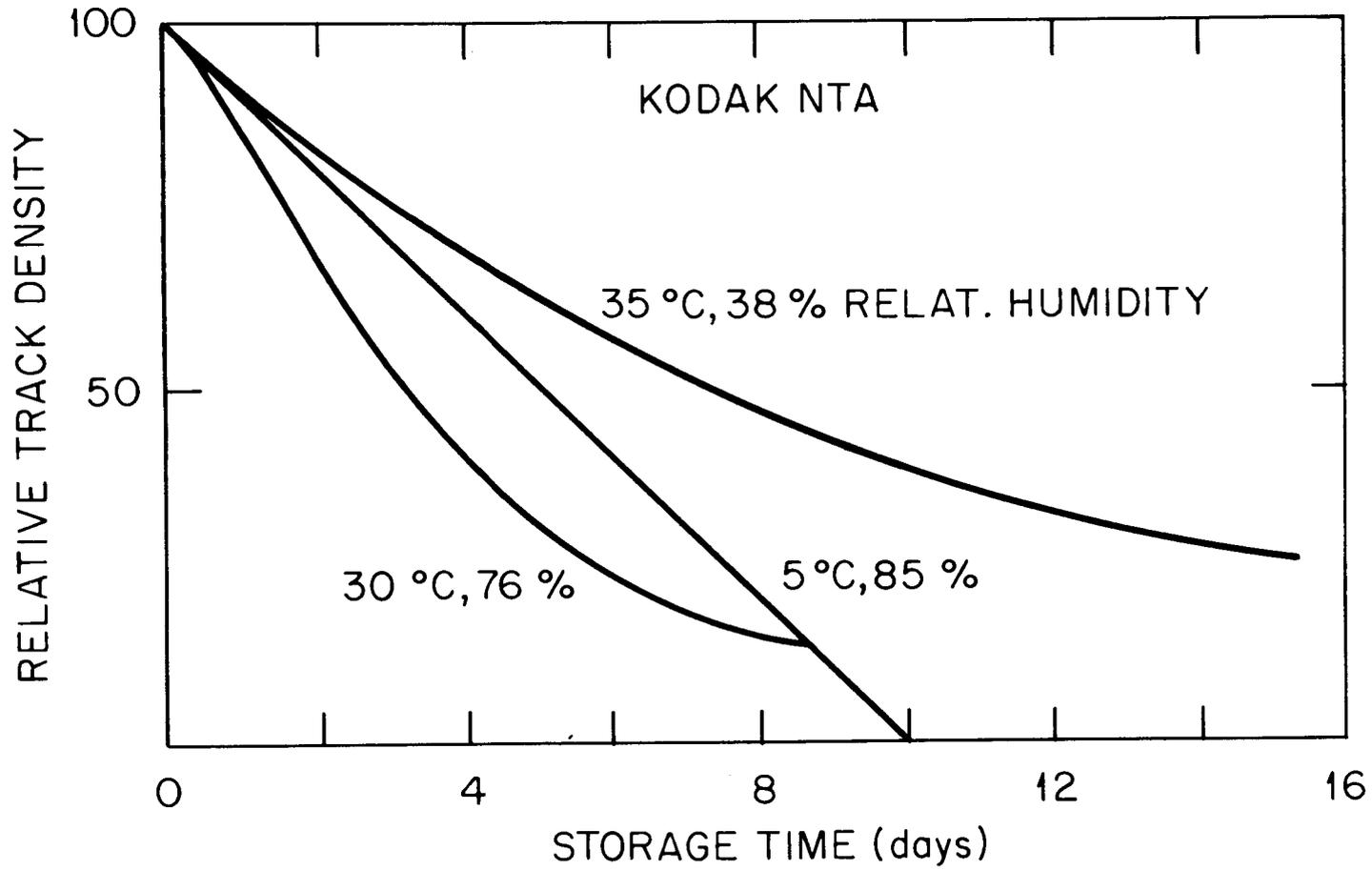


Fig. 9 Relative track density in Kodak NTA film for storage at different temperatures and humidities (after 29).

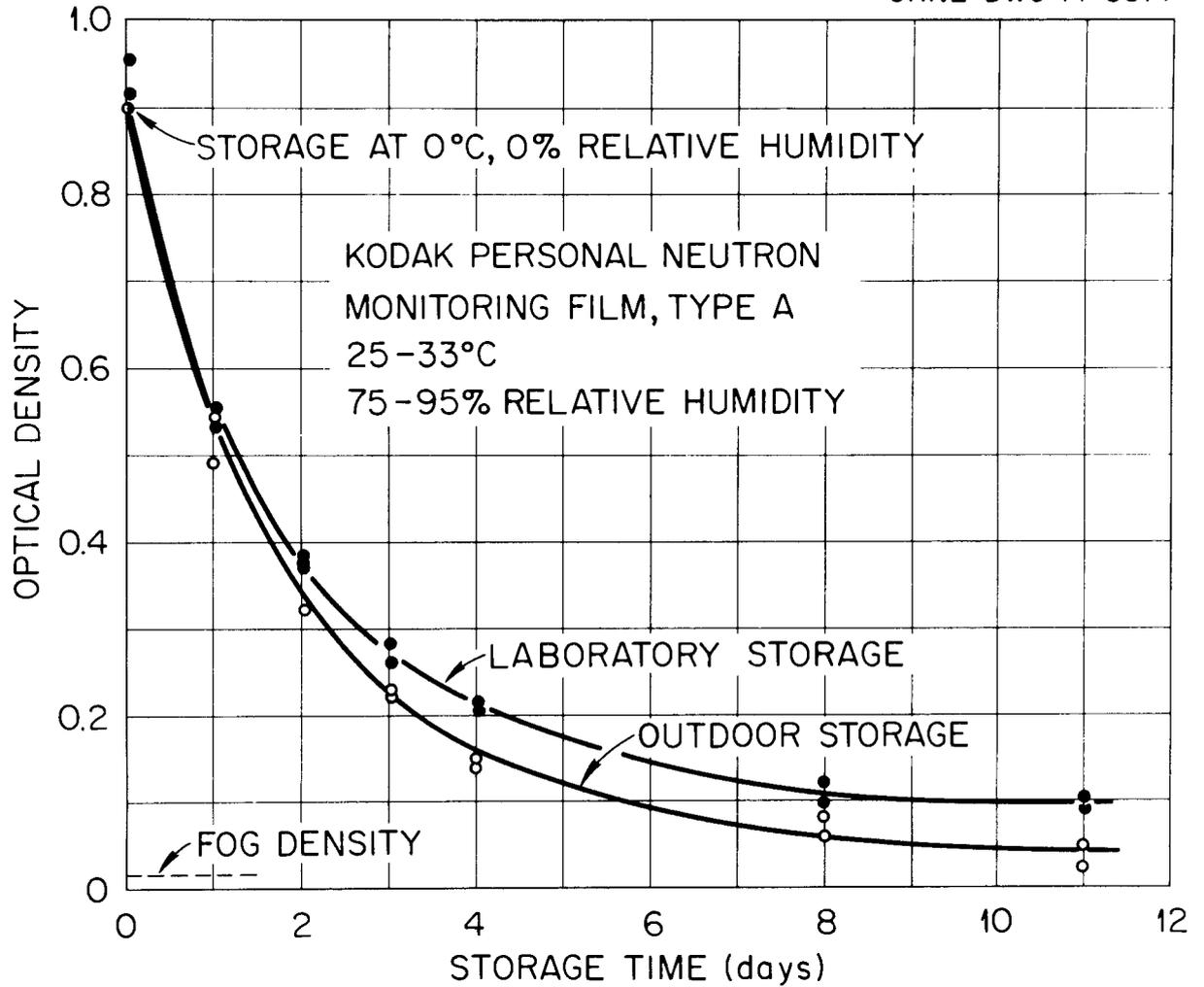


Fig. 10 Fading of latent image in a Kodak NTA film in a semi-tropical climate.

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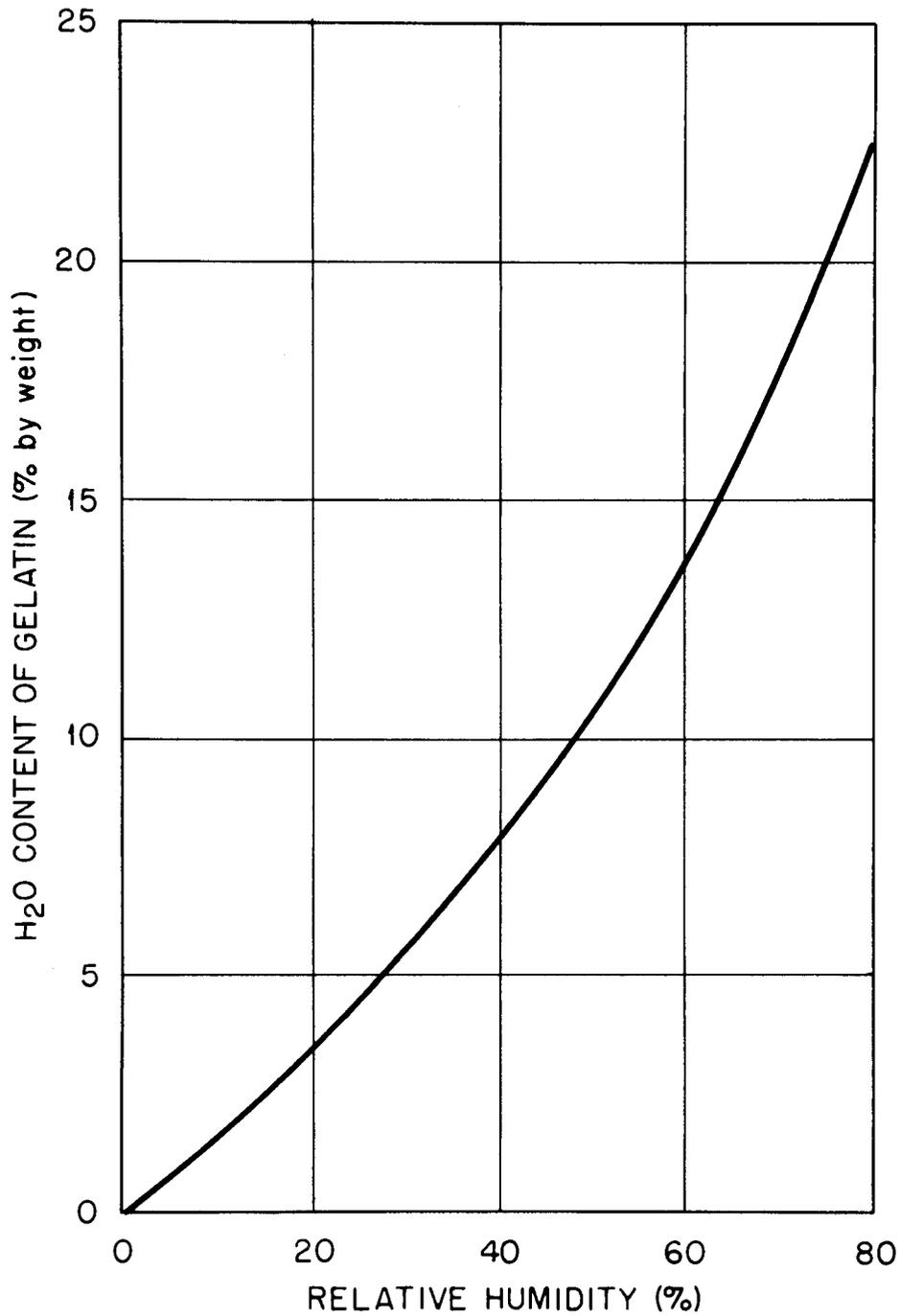


Fig. 11 Water content of the gelatin in a nuclear track emulsion at 20°C as a function of relative air humidity (after 33).

Table 1. Permeability of Some Polymers to Water Vapor at 25°C

Polymer	$P \times 10^{10}$ *)
Ethyl cellulose	12,000
Cellulose nitrate	6.300
Cellulose acetate	5.500
Polycarbonate	1.400
Polystyrene	1.200
Polyvinyl chloride	156
Polyethylene terephthalate	130
Polyethylene, low density	90
Polypropylene	51
Polyethylene, high density	12
Polyvinylidene Chloride	0.5
Polytrifluorochloro- ethylene, amorphous	0.29

\*) Permeability coefficient, defined as the produce of the amount of permeant and the film thickness, divided by the product of area, time, and pressure-drop across the film.

humidity outside, at an increased ambient temperature (which are the conditions to be found in a tropical climate). Indeed, field experiments (Fig. 5) shows little difference in the fading of Kodak Type 2 films with and without sealing in thin "protective" polyethylene bags. Sufficiently thick plastic may, of course, retard the humidity more efficiently. In Fig. 12, the fading rate in an equilibrated Kodak NTA film in the factory-provided wrapping is compared to that of such a film in a special thick polyethylene holder, with and without previous drying.

The relatively most efficient vapor barrier is a plastic-aluminum-paper compound ("pipe tobacco pouch paper"), in which the metal foil acts as the actual barrier, and the polymer is used only for heat-sealing with a wide ( $\sim 5$  mm) rim. The Kodak NTA film should only be used for short ( $\sim 1$  week) monitoring periods in a very dry and cool climate unless protected by such a sealing, which is commonly used by the photographic industry to protect their fresh film packs. Even with the pouch paper sealing, however, NTA films are by no means fully protected against fading. In one experiment, films were heat-sealed immediately after being removed from the freshly opened factory packaging, equilibrated for 12 days at  $30^{\circ}\text{C}$  and 95 % relative humidity, and then exposed to fast neutrons from a Pu/Be source (behind 2 cm of Pb) at different times before processing one month after sealing. As can be seen in Table 2, after one week of storage about one third, after two weeks two thirds, and after three weeks all the tracks have disappeared under these rather realistic conditions.

Only one photographic dosimeter is commercially available with pouch-paper type of packaging for the individual film. This is the Kodak-Pathé

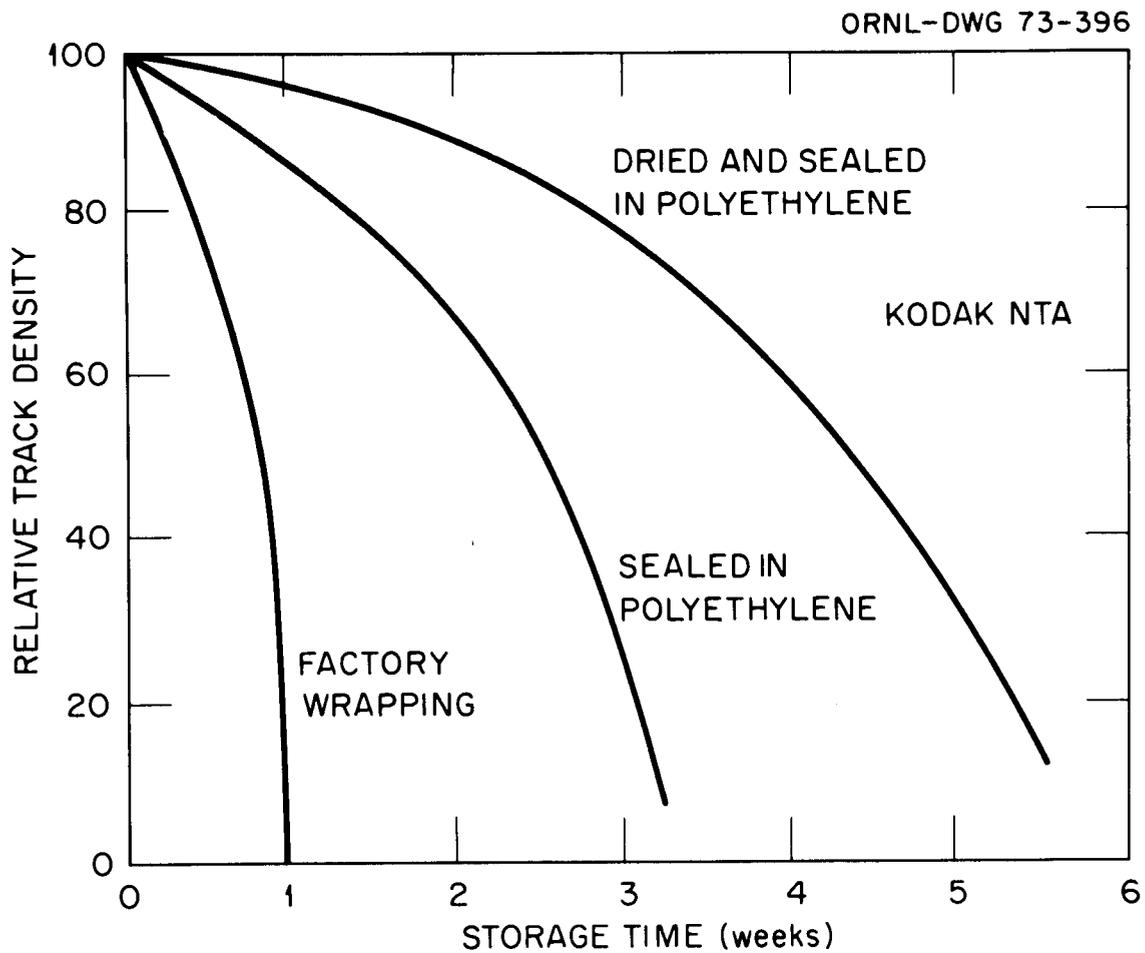


Fig. 12 Track fading in Kodak NTA film at ambient temperature and 100 % relative humidity in normal wrapping; placed in a protective thick polyethylene holder; and with both film and holder carefully dried before sealing (after 28).

Table 2: Effect of "Pouch Paper" Sealing on Residual Track Density (in Percent)  
in Kodak NTA Film

storage time (weeks)	unsealed			
	0 % rel. humid., -10°C	95 % rel. humid., 30°C *)	0 % rel. humid., -10°C	95 % rel. humid., 30°C *)
1	100	0	100	78**)
2	100	0	100	37**)
3	100	0	100	0

\*) Films equilibrated for 12 days after removal from freshly opened package at given climate prior to first exposure.

\*\*\*) Many of the tracks thin and difficult to see

Type 1, which is not a nuclear track film, but a paper base on which different emulsions for X- and  $\gamma$ -radiation dosimetry (to be evaluated by reflection densitometry) are coated. It is, incidentally, widely assumed that in the fresh dosimeter packs as received from Kodak the films have been desiccated prior to packaging and sealing. In fact, the films are sealed after being equilibrated at 21°C and 40-50 % relative humidity and should, therefore, be stored in a desiccator before any additional sealing is carried out (the British National Radiation Protection Service provides for use in tropical countries films which have been sealed in plastic-aluminum-paper laminate; usability for up to two months is claimed for these films). One problem with pouch-paper sealing is that the films become too large for the common film badges, which makes it necessary to either redesign the badge, or to use smaller dosimeter films. In addition, new calibration curves for the filter-analytical evaluation may be required due to the effect of the additional absorber layers on the observed film densities.

In a recent pilot study which was carried out for the Pan American Health Organization, Regional Office of the World Health Organization, in cooperation with the national authorities, various types of films as used by the national personnel monitoring services have been stored together with TLD powders for periods of  $\sim$  2 to 3.5 months in a number of locations in four countries with rather different climates (Table 3). Some of the films were stored partly unexposed in order to study changes in the background (fog) density, and some were stored after on-location exposure to gamma radiation (radium or  $^{60}\text{Co}$ ) doses in the 0.4 to 2 rad range. Irradiations were carried out in a low-scattering environment

Table 3: Storage Conditions of Dosimeters During South American Pilot Study

Country	City	Period of Storage	time of storage (months)	Climate During Storage Period			
				Average temperature (°C)	maximum temp. (°C)	average humid. (°C)	maximum r. h. (%)
Colombia	Bogota	Dec. 1-March 1	3	16		70	
	Cartagena	Dec. 1-March 1	3	28		90	
Chile	Santiago	Dec. 1-Feb. 29	3	20.7	34.2	71	87
	Los Angeles	Dec. 1-Feb. 29	3	18.6	38.2	63	78
Ecuador	Guayaquil	Dec. 1-March 15	3.5	29	32	82	95
Argentina	Buenos Aires	Dec. 17-Feb. 15	2	29.0	34.8	88	100
	Santa Fe	Dec. 20-Feb. 13	1.9	31.0	39.7	78	100
Control	Oak Ridge	Dec. 22-April 73	5	-8	25**)	0	0

\*) During the period not stored on the indicator location, the dosimeters were either being mailed (5-10 days), or stored together with the controls at -10°C, 0 % rel. humid. at ORNL.

\*\*\*) Short periods during travel (5-15 days), otherwise over CaCl<sub>2</sub> in deep-freezer at ORNL.

under electron equilibrium conditions, but without additional filters. Half of the films were individually heat-sealed, mostly immediately after being removed from the factory package, but without additional dessication. The sealing was carried out with a soldering iron, in a wide-rimmed envelope consisting of a laminated compound Al-plastic "pouch paper" as used by the photographic industry for protecting larger packs of X-ray and dosimeter films. The following film types were used in some, or all, of the storage tests:

1. Kodak Personnel Monitoring Film Type 3, Emulsion No. M 330 22701;
2. Kodak PM Film Type 2, Emulsion No. 205 702; and
3. Agfa/Gevaert Personnel Monitoring Film.

When not being stored in a normal office or laboratory room (protected from water, direct sunshine and radiation sources, but not air-conditioned) at the listed locations, the films were either under uncontrolled conditions in the mail for periods of 5 to 10 days, or stored in a polyethylene container over dry calcium chloride in a deep-freezer at the ORNL DOSAR Facility. It had been shown in previous tests that fading is negligible under these latter conditions. The films were processed jointly on April 25, 1973 by the ORNL film dosimetry service with the standard development for Kodak PM films, and read the following day with an Ansco-Macbeth Densitometer by averaging several optical density measurements at different locations on each film. Each number given in Tables 4 and 5 and discussions represents the average readings of at least two films. The agreement between identically treated films was usually excellent, with the exception of some sealed films which had been stored in severe climates. In this case, microscopic imperfections in

Table 4: Net Optical Density of Gamma-Irradiated Films, in Percent of Controls

Location	Storage time (months)	Kodak Type 2		Kodak Type 3			Agfa-Gevaert		
		sensitive emulsion		sensitive emulsion	insensitive emulsion	sensitive emul.			
		unsealed	sealed	unsealed	sealed	unsealed	sealed		
Bogota, Colombia	3	97.5	97.5	108	86	~ 75	~ 75		
Cartagena, Colombia	3	21.3	46.3	19.2	91.3	~ 90	~ 100		
Guayaquil, Ecuador	3.5	0	11.3	7	105,11*)	~ 15	~ 15		
Santiago, Chile	3	103.5	97.6			~ 100	~ 100		
Los Angeles, Chile	3	102.9	96.5			~ 100	~ 100		
Buenos Aires, Argentina	2	29	92	< 25		~ 100	~ 100	78.3	41.0
Santa Fe, Argentina	1.9	67	97.5			~ 100	~ 100	83.3	72.4

\*) Two films, one evidently with imperfect sealing

Table 5: Fog Optical Density of Unexposed Films

Location	Kodak Type 2		Kodak Type 3				Agfa-Gevaert			
	sensit. em.		sensit. em.		insensit. em.		sensit. em.		insensit. em.	
	unsealed	sealed	unseal.	seal.	unseal.	seal.	unseal.	seal.	unseal.	seal.
Bogota	0.33	0.29	0.41	0.42	0.08	0.8				
Cartagena	0.27	0.23	0.33	0.19	0.11	0.11				
Guayaquil	0.25	0.16	0.16	0.15	0.12	0.11				
Santiago	0.34	0.32	0.48	0.47	0.07	0.08				
Los Angeles	0.30	0.30	0.42	0.42	0.08	0.08				
Buenos Aires	0.18	0.29		0.42	0.07	0.08	0.26	0.29	0.10	0.10
Santa Fé	0.26	0.30			0.07	0.07	0.18	0.16	0.10	0.10
Control (Oak Ridge)	0.29		0.40		0.07		0.30		0.16	

the sealing produced some large differences in the O.D. of the films. A few films turned out to be over- or underexposed for exact evaluation, or stuck to the film wrapping. Best estimates, or no values at all are given in these cases.

The results are summarized in Table 4. Corrections have been made for variations in the fog densities which may increase or (more frequently) decrease, depending on conditions at the locations where the films were stored. This is caused by the superimposed effects of fading, fogging, and accumulation of background radiation ( $\sim 30$  to  $80$  mrad, depending on the location - the DOSAR storage area being on the higher side because of the nearby unshielded HPRR reactor). As can be seen in Table 5, which includes the measured fog densities in films which have not been exposed to any but background radiation, fading of the fog amounts to up to 45 % in the Kodak Type 2 sensitive emulsion stored in Guayaquil, and up to 63 % in the sensitive emulsion of Kodak Type 3. In other locations such as Bogota and Santiago, fog build-up by up to  $\sim 20$  % dominates, to be explained at least in part by the fairly high background at these locations (a total of 64 mR in Santiago and 42 mR in Bogota, as measured with  $\text{CaSO}_4:\text{Dy}$ ). Apparently the insensitive emulsions show less fading and more fogging.

This tendency is also evident in the exposed films, where the fading in films stored at four locations (Cartagena, Guayaquil, Buenos Aires, and Santa Fé) was more pronounced in the sensitive than in the insensitive emulsions. In these four locations, the fading in, for instance, the sensitive emulsion of the unsealed Kodak Type 2 film amounted to at least one third (in Santa Fé), and a maximum of 100 % fading in Guayaquil.

*In these cities - more than one half of all the locations in this study- Kodak Type 2 and 3 films are clearly not suitable for long-term (one month or more) radiation monitoring.* In three other locations with relative low average temperatures and humidities, namely Bogota, Santiago and Los Angeles, however, the observed stabilities are apparently sufficient for monitoring periods of up to three months.

The as-described optimum (or near-optimum) sealing had in most cases a beneficial effect on film stability, both with regard to the fog and the radiation-induced effect. Even in the carefully sealed Kodak 2 film, however, fading amounted to 89 % in Guayaquil and 54 % in Cartagena. Despite such sealing, films should not be used in two out of the seven locations. In Fig. 13, the appearance of Kodak Type 2 films is given after exposure to the same radiation dose and storage in Bogota, Cartagena, and Guayaquil, with and without sealing. The striking differences in the optical density are clearly visible.

In the Agfa-Gevaert films stored in Buenos Aires and Santa Fé, Argentina, the fading was even higher in the sealed than in the unsealed films. In this case, the films had not been taken from a freshly opened package for sealing, but had equilibrated for some time with the ambient humidity. The sealing had evidently prevented the films more from becoming dry than from becoming moist, demonstrating the importance of careful dessication just prior to sealing in a dry room.

### 3. Thermoluminescence Phosphors

The slow, thermally stimulated release of trapped electrons or holes in irradiated TLD materials has been described in terms of trap depth (activation energy) and frequency factor, based either on the classical

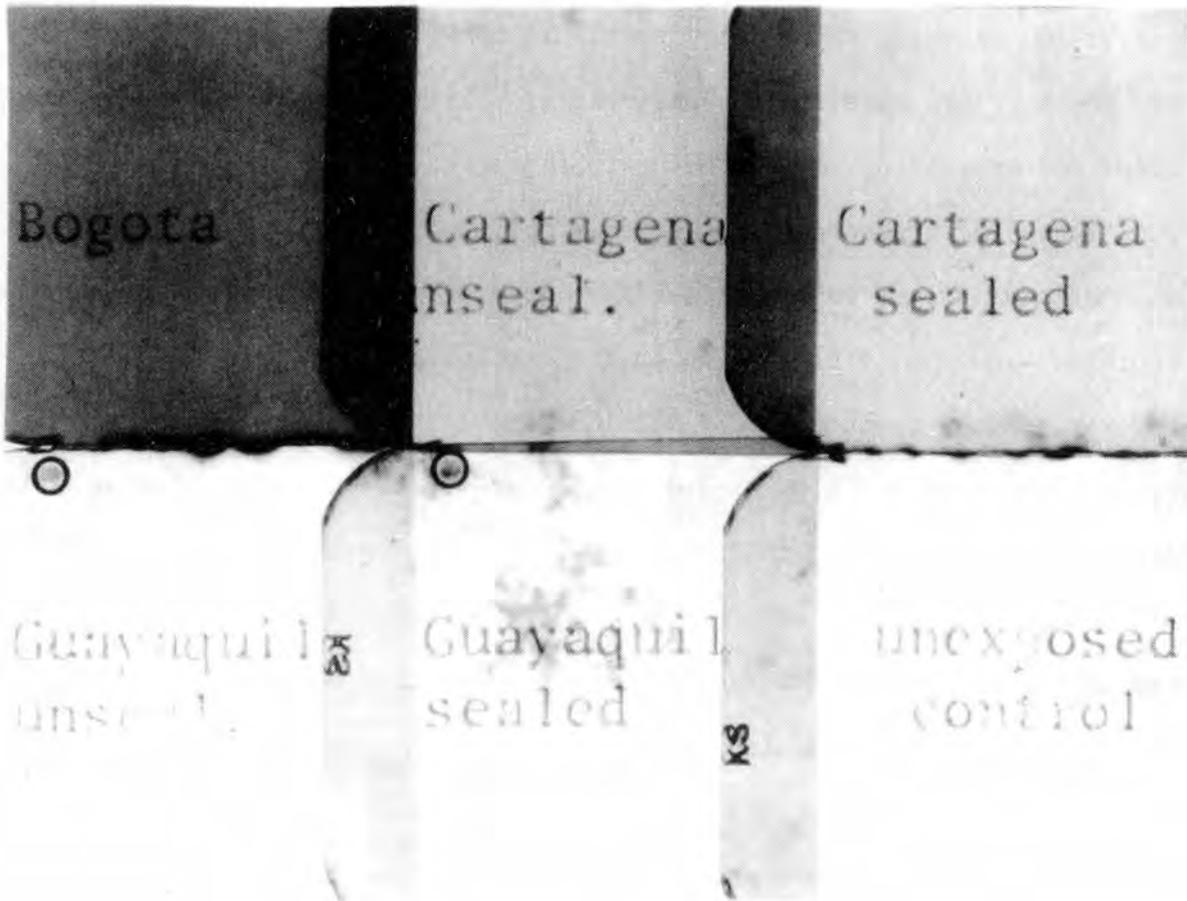


Fig. 13 Kodak PM Type 2 films uniformly irradiated at the same time to 0.4 R gamma radiation and stored for about three months

- a. unsealed in Bogota, Colombia;
  - b. unsealed in Cartagena, Colombia;
  - c. sealed in Cartagena;
  - d. unsealed in Guayaquil, Ecuador; and
  - e. sealed in Guayaquil.
- On the lower right side, unexposed control film.

Randall-Wilkins equations, or on other slightly more sophisticated models (for a recent compilation, see ref. 1). The reliability of the calculated fading predictions depends mostly on the precision with which the trap parameters of a given phosphor are known, and on the assumed TL mechanism. As a rule, the thermal stability of the radiation-induced "latent" signal in a TL phosphor increases rapidly with increasing temperature of the peak location. A peak location of 100°C roughly corresponds to a half-life of the trapped electrons or holes at 20°C of around 3 hours; for a 200°C peak, the half-life has increased to about a year; at 300°C it is  $\sim 3000$  years, and at 400°C,  $\sim 10^7$  years.

Disagreement between the values calculated from experimental trap parameters and the actual fading results may be due to experimental errors in either measurements, but are explainable in other phosphors with the presence of complicating secondary processes including re-trapping between different trapping levels, trap transformations, thermal quenching, and "abnormal" fading due to non-radiative recombination processes. In no case should calculated fading rates be assumed to be correct without verification. This is also true for the temperature dependence of the fading rate. Although it is safe for some phosphors to assume a simple linear extrapolation of fading rates which have been measured at elevated temperatures down to lower temperatures as indicated in Fig. 14, such an extrapolation is not possible in many other cases. In one modern highly sensitive and stable phosphor, for example, there is an actual increase in the TL signal during storage at lower temperatures, probably caused by re-trapping from deeper traps into the TL traps (Fig. 15).

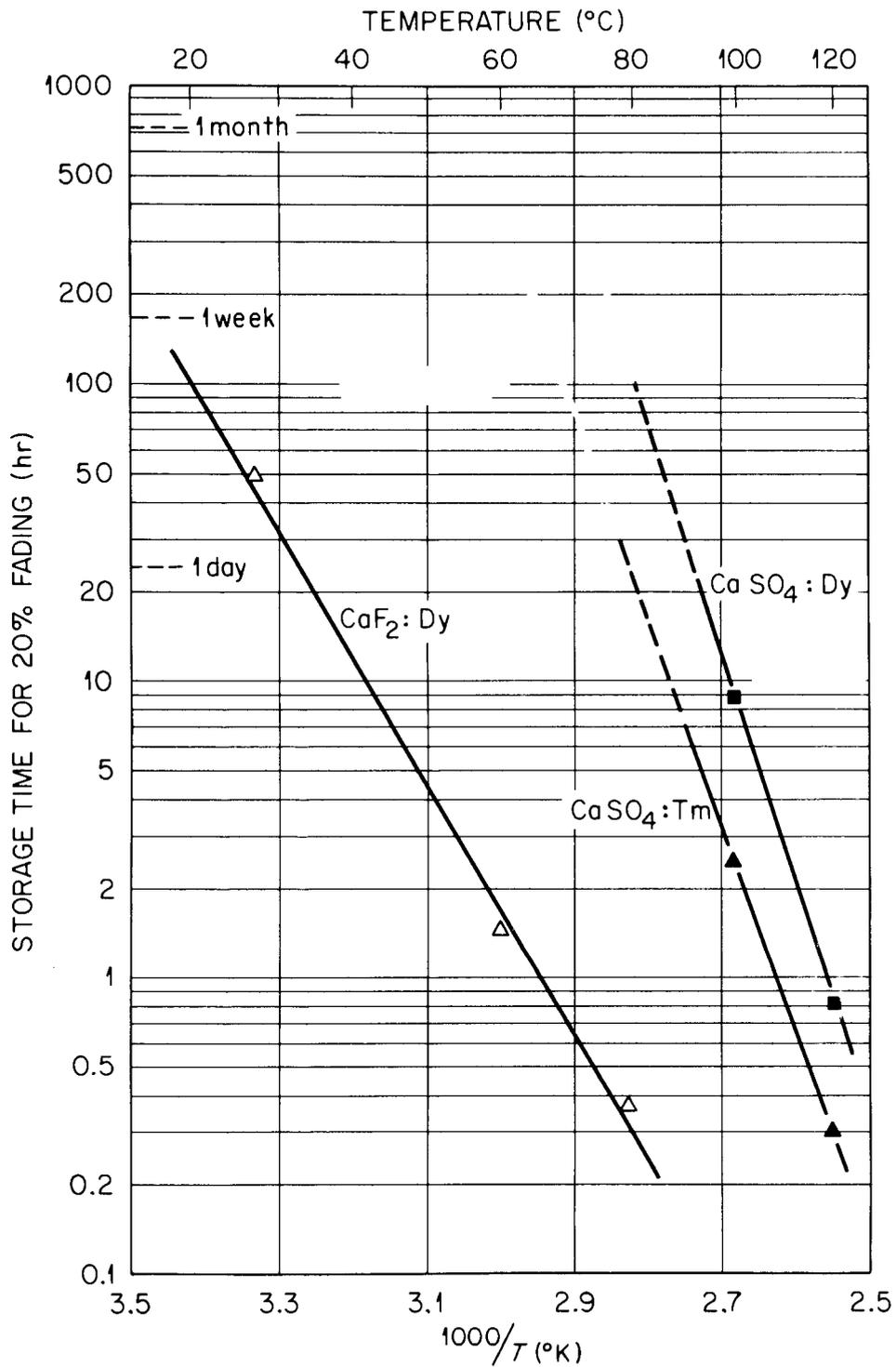


Fig. 14 Simplified diagram of the effect of storage temperature on the fading rates in three sensitive TL phosphors.

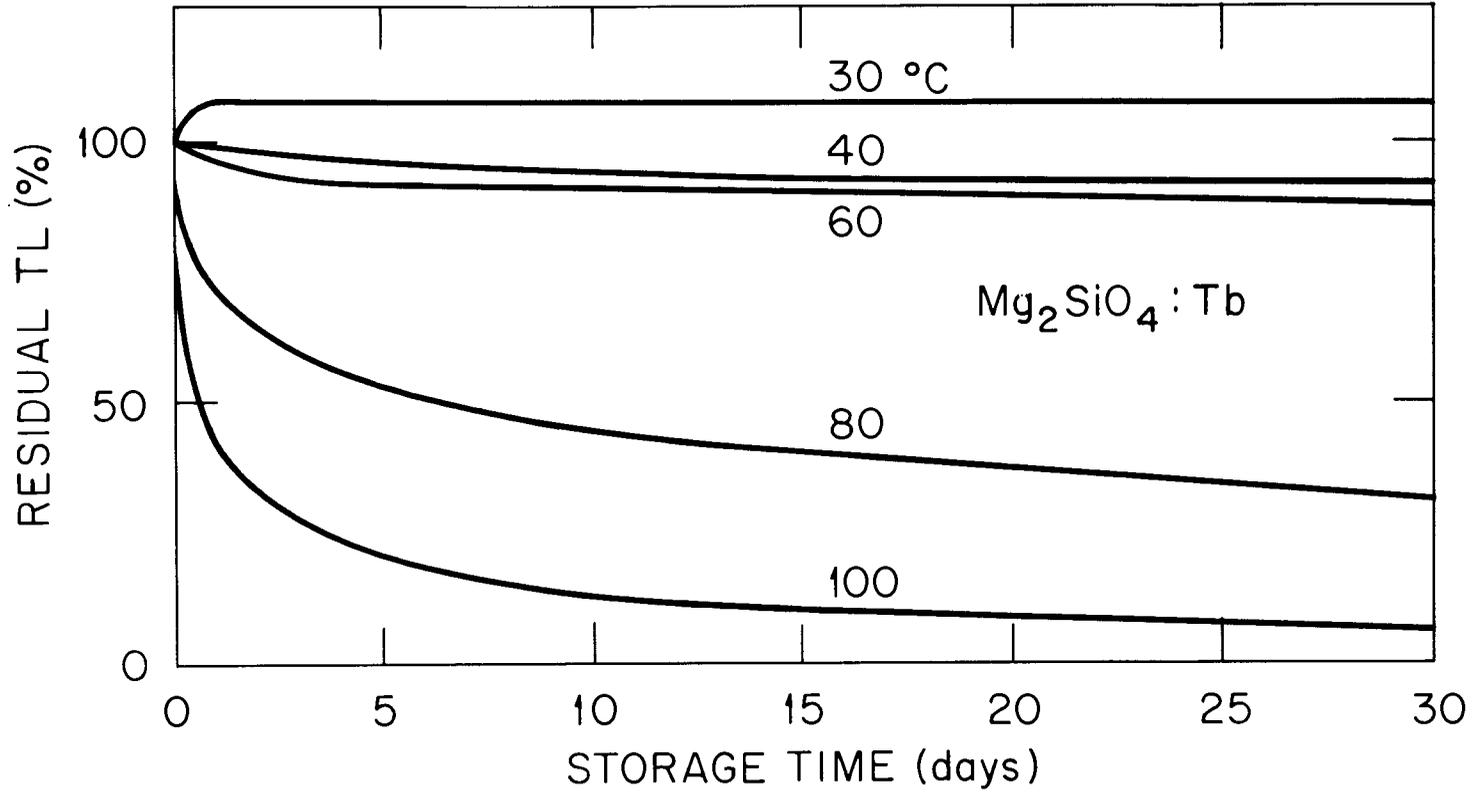


Fig. 15 Fading and build-up of the TL signal in  $Mg_2SiO_4:Tb$  at different storage temperatures (after 80).

In another widely used phosphor,  $\text{CaF}_2:\text{Mn}$ , 6-10 % of the signal faded during the first 16 hours of storage in the dark at room temperature despite a high peak temperature ( $\sim 260^\circ\text{C}$ ) in this material. The amount of fading depends on the heating rate. No fading was observed at very low rates around  $1^\circ\text{C}/\text{sec}$  (there is a strong effect of the heating rate also on the apparent sensitivity). These effects are explained by preferential thermal quenching of the more stable components of the TL glow-curve (88). If read in one modern system developed for the U.S. Navy,  $\text{CaF}_2:\text{Mn}$  exhibits an "abnormal fading" of 2.4 % per decade of time independent of the storage temperature between 20 and  $100^\circ\text{C}$  (94). In some natural  $\text{CaF}_2$  as well as commercial  $\text{CaF}_2:\text{Mn}$  phosphors, self-dosing due to the radioactivity of the dosimeters complicates precise low-dose measurements and fading studies (89,90).

Some phosphors such as  $\text{CaSO}_4:\text{Mn}$  are clearly not suitable for our purposes because of their rapid fading (Fig. 16). Others such as  $\text{CaF}_2:\text{Dy}$  have a wide trap depth distribution and also exhibit rapid fading (about 20 % during 10 hours at  $20^\circ\text{C}$  according to ref. 94 - see also Fig. 17), but may be considered for personnel or environmental monitoring if they are evaluated after a "stabilizing" heat treatment (such as 10 min. at  $115^\circ\text{C}$ ) which anneals all the low-temperature traps. It is not the purpose of this report to review all the TL fading data which can be found in the literature. Instead, it will focus on a few phosphors which either are widely used such as  $\text{LiF}:\text{Mg,Ti}$ , (manufactured by the Harshaw Chem. Co., Cleveland, Ohio, under the name TLD-100), or which are promising with regard to higher sensitivity and stability.

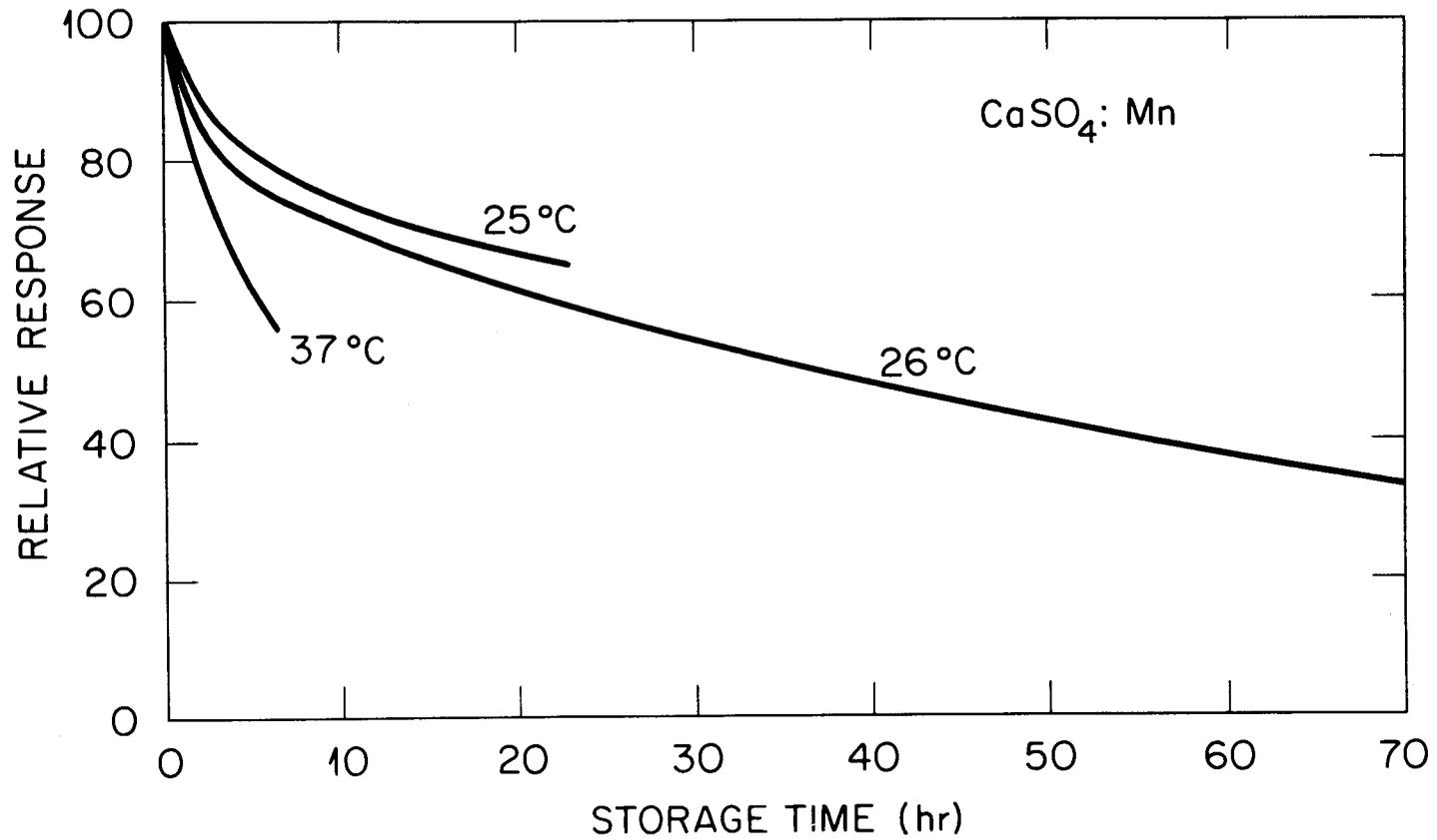


Fig. 16 Fading of TL in CaSO<sub>4</sub>:Mn at 25 and 37°C (after 81), and at 26°C (after 82).

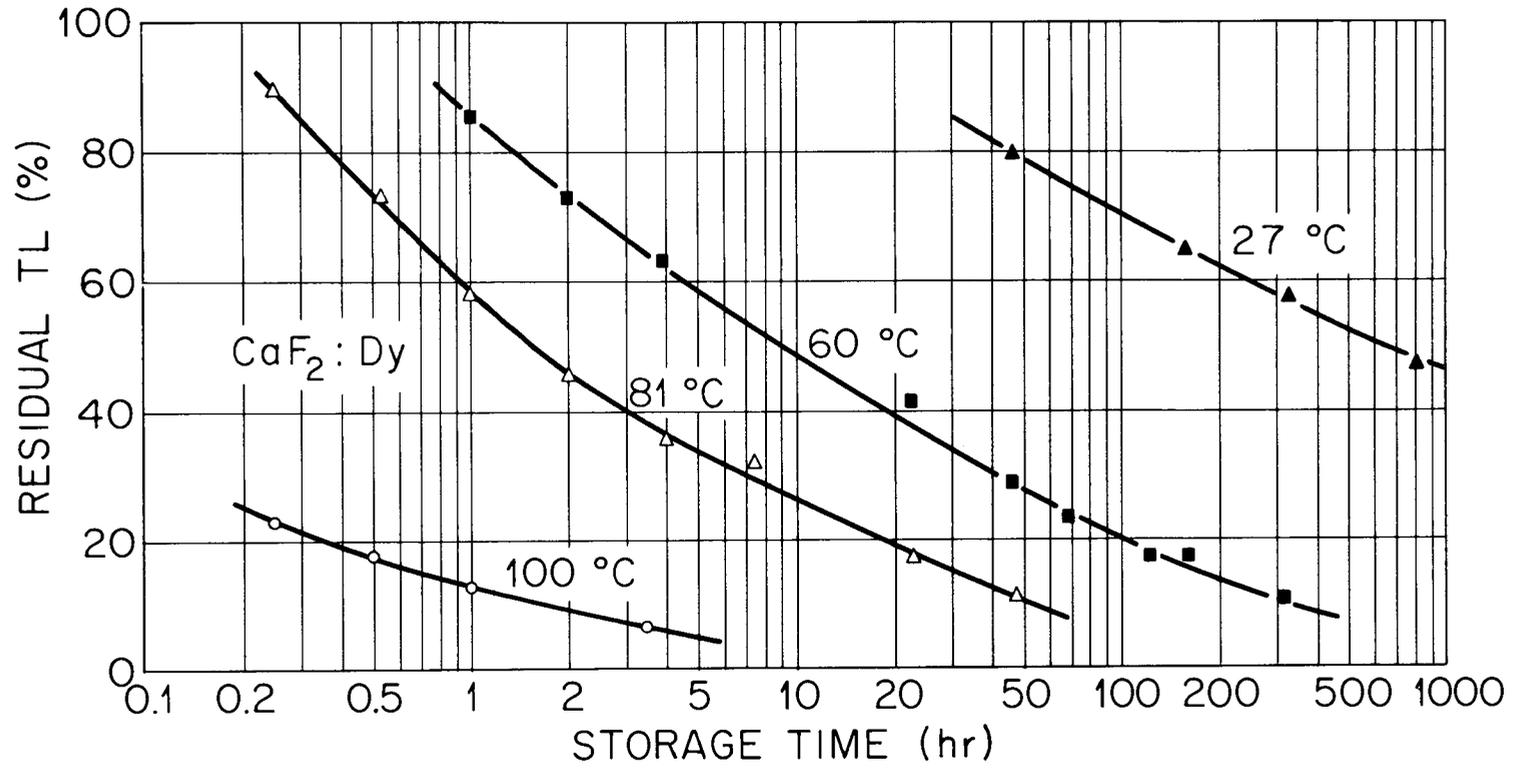


Fig. 17 Fading of TL in CaF<sub>2</sub>:Dy at different storage temperatures.

Humidity is known to have little effect on the storage stability (fading, sensitivity, background) of all but a few hygroscopic TL phosphors such as  $\text{Li}_2\text{B}_4\text{O}_7:\text{Mn}$ . Only in  $\text{LiF}:\text{Mg},\text{Ti}$  powder, a strange increase in the TL signal has been observed by one group (6) if the irradiated phosphor was stored for 10-30 days at 95-100 % relative humidity. Exposure to intense light, in particular UV, can induce as well as anneal TL signals in most phosphors, the amount of change depending on the pre-exposure level, spectrum of the light, and phosphor. The phosphors in our experiments have, therefore, been protected from intense light by encapsulation in small plastic vials (not glass, whose  $^{40}\text{K}$  content results in a dose build-up during extended storage) which were placed in a non-transparent paper or plastic wrapping. Handling and readout was done in normal natural or artificial laboratory light.

The detectors were annealed just prior to irradiation either by simply heating them for  $\sim 30$  min. to  $\sim 400^\circ\text{C}$  ( $\text{Mg}_2\text{SiO}_4:\text{Tb}$ ,  $\text{CaSO}_4:\text{Dy}$ ,  $\text{CaSO}_4:\text{Tm}$ ), or with the standard annealing treatment for  $\text{LiF}:\text{Mg},\text{Ti}$ , consisting of one hour at  $100^\circ\text{C}$ , cooling at a constant rate, and  $\sim 24$  h at  $80^\circ\text{C}$ . Each data point represents the average of 5-10 readings from the same vial, dispensed by a standard vibrator attached to the Harshaw 2000 reader system. The reading was done at optimized temperatures for each phosphor in a constant pure nitrogen flow. Two minutes after the end of a heating cycle, a second heating of the same sample established the "background" to be subtracted from the signal. The standard deviation of the readings within each set amounted to 2-3 %.

The results of one of our recent sensitivity and stability comparisons of some TLD phosphor powders are compiled in Table 6. As can be seen, several phosphors are as stable (or more stable) and 20 to 30 times as sensitive as LiF:Mg,Ti which has, however, the advantage of a rather flat photon energy response. This may indeed be an important factor for personnel dosimetry in medical radiology, but it has been shown (30,31) that energy independence is not important for environmental dosimetry, because the contribution of photon energies  $< 100$  keV to total dose is negligible under almost all practical conditions. In intercalibration studies, at least of high-energy photon and electron sources, the detector(s) do not have to be energy-independent either, because some shielding easily protects them from low-energy scattered radiation.

Even if one ignores some obviously erroneous early results, the data in the literature on the fading in LiF:Mg,Ti are not consistent. Extrapolation of measurements at higher temperatures ( $100^{\circ}\text{C}$ ) down to lower temperatures would result in very low fading rates (one calculated value is  $\sim 3$  % fading during two months at  $50^{\circ}\text{C}$ ). Actually, higher rates have been observed, ranging from  $\sim 5$  % during three months at  $\sim 22^{\circ}\text{C}$  (34,35), to  $\sim 12$  % during six weeks at  $32^{\circ}\text{C}$ ; and  $\sim 10$  % during one day at  $60^{\circ}\text{C}$  in the author's studies (Fig. 18). There are two main reasons for these inconsistencies:

1. as there are rapidly fading low-temperature peaks in LiF:Mg,Ti, the measured fading rate depends on the type of read-out, being less pronounced in modern readers with a pre-heating cycle which excludes those peaks from the light integration (another method to prevent

Table 6: Relative Sensitivities and Stabilities in Some TLD Phosphors

Phosphor	Relative Sensitivity *)	Change during 6 weeks at 32°C (%)
LiF:Mg,Ti	1	-10
CaF <sub>2</sub> :Dy	15	-20 to 60 **)
CaSO <sub>4</sub> :Tm	22	-12
CaSO <sub>4</sub> :Dy	23	-1.5
Mg <sub>2</sub> SiO <sub>4</sub> :Tb	30	+8

\*) const. phosphor volume, Harshaw 2000 reader

\*\*.) depending on pre-readout temp. treatment

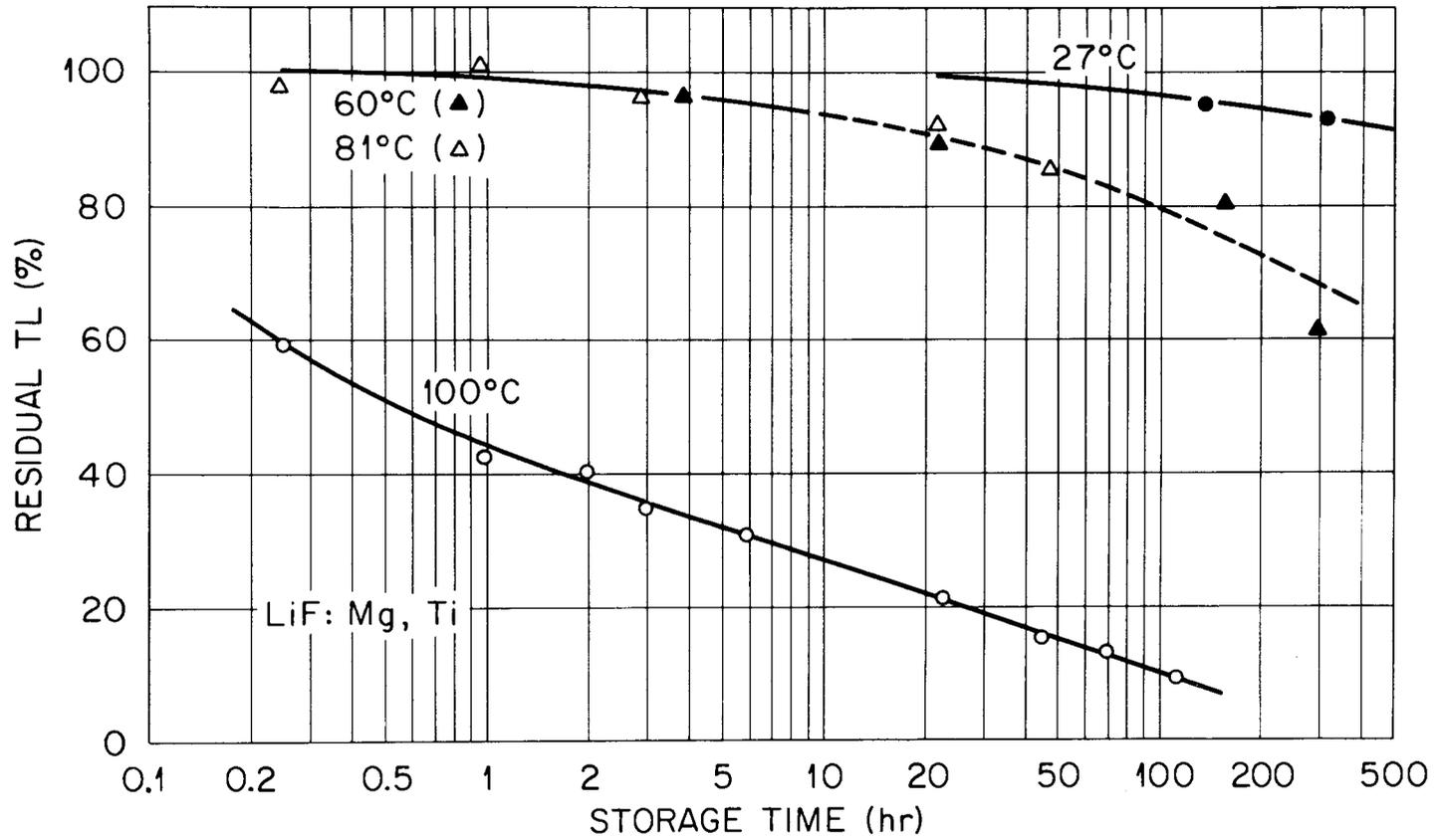


Fig. 18 Fading of TL in LiF:Mg,Ti (TLD-100) powder at different storage temperatures.

reading of these peaks is to keep the irradiated phosphor for  $\sim 30$  min. at  $\sim 80^\circ\text{C}$  prior to the read-out; this reduces the signal by  $\sim 5\%$  as compared to a freshly irradiated LiF:Mg,Ti sample);

2. complex trap transformations and/or re-trapping processes occur during storage, resulting in slow changes in both the height and the area of various TL peaks which are accelerated by temperature increases (36). It has been suggested (93) to reduce the fading, which is observed in particular in LiF:Mg,Ti samples which did not undergo the usual annealing cycle (1 h at  $400^\circ\text{C}$ , 24 h at  $80^\circ\text{C}$ ), to use special readout techniques, such as adding peak-height and glow-curve area readings. There also may be some "abnormal" non-radiative trap leakage fading.

In one of the most interesting of the modern phosphors, namely  $\text{CaSO}_4:\text{Dy}$ , only a little fading ( $\sim 1-2\%$  during one month,  $\sim 5-8\%$  during six months) has been detected at room temperature ( $\sim 22^\circ\text{C}$ ). The fading rate increases slightly to about  $3\%$ /month at  $30^\circ\text{C}$ . For the fading rate at higher temperatures, see Fig. 19. There have been a few reports on higher fading rates, amounting to as much as  $30\%$  during the first month of storage at  $25^\circ\text{C}$ , and another  $\sim 8\%$  during the following six months, in one self-prepared material (37). At least one of these reports was shown since to have been erroneous. In another, a residual low-temperature peak around  $100^\circ\text{C}$  was apparently more pronounced than in the commercial product. In a more recent study (6), the same group reports no fading in the  $\text{CaSO}_4:\text{Dy}$  manufactured by Harshaw after up to three months at  $25^\circ\text{C}$  and relative humidity up to  $100\%$ , but also  $\sim 20\%$  fading during one month at  $35^\circ\text{C}$  or  $\sim 12\%$  during 3 months at  $25^\circ\text{C}$ , in apparently the same product.

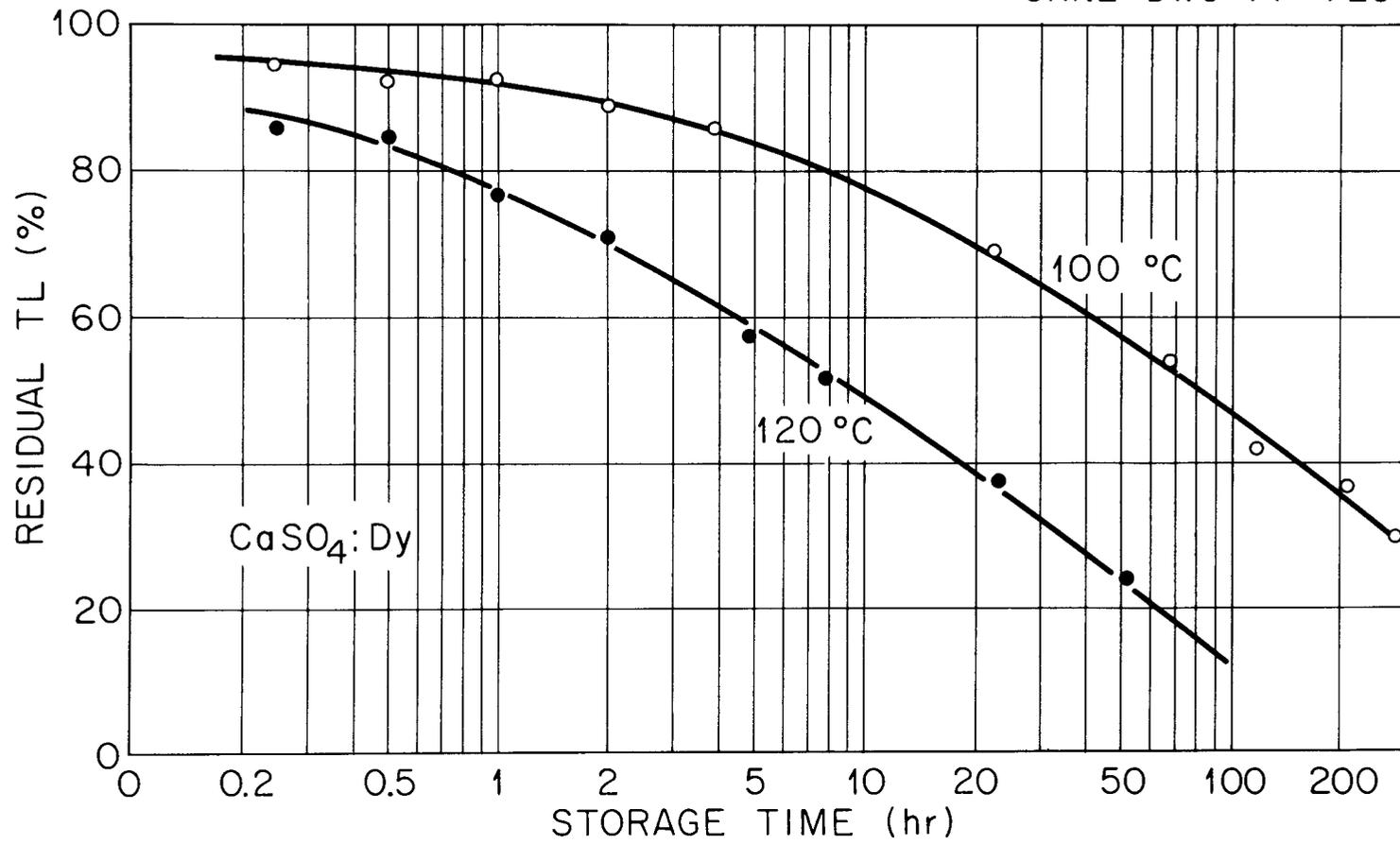


Fig. 19 Fading of TL in CaSO<sub>4</sub>:Dy powder at different storage temperatures.

In the Latin American pilot study (the results are summarized in Table 7), the LiF:Mg,Ti exhibited (as expected on the basis of our previous laboratory experiments) at all locations more fading than the CaSO<sub>4</sub>:Dy. In Santiago and Los Angeles, there was a slight increase in the reading of the CaSO<sub>4</sub>:Dy, explainable at least in part by higher background radiation levels in these locations (64 and 47 mR, respectively, compared to 30-40 mR in Cartagena, Buenos Aires and Bogota). The maximum observed fading was 23 % in LiF:Mg,Ti (in Cartagena), and 5.4 % in CaSO<sub>4</sub>:Dy (in Bogota and Guayaquil).

If one considers 20-30 % a tolerable maximum fading rate in personnel monitoring, the two phosphors would be suitable for this purpose in all locations. For intercomparison studies with its higher accuracy requirements, and for environmental monitoring with higher possible temperatures (direct sunshine on the stations) CaSO<sub>4</sub>:Dy is clearly superior to LiF:Mg,Ti for reasons of stability as well as sensitivity. Even with this material, however, it is advisable to include a sample which has been exposed to a high, known radiation dose as a fading indicator in each station or detector package (the reading of this sample is compared to that of an identical sample stored under low-fading conditions in a deep-freezer for correction of the error due to fading). The results of this study agree well with those of similar field tests previously performed by the author in Taiwan (38), and unpublished environmental radiation measurements at ORNL and in private homes in the Oak Ridge area.

Table 7: Thermoluminescence of LiF:Mg,Ti (TLD-100) and CaSO<sub>4</sub>:Dy During South American Pilot Study in Percent of Controls\*)

Location	LiF:Mg,Ti	CaSO <sub>4</sub> :Dy
Bogota	89	94.7
Cartagena	77.2	97.6
Guayaquil	86.2	94.6
Santiago	87.5	106
Los Angeles	92.7	109.5
Buenos Aires	90.5	100
Santa Fe	82.6	97

\*) Stored together with films for the same time and under identical conditions, as specified in Table 2; powdered phosphors heatsealed after annealing and prior to on-location irradiation to 0.4-2 R gamma radiation in medical polyethylene tubing provided by Clay Adams, Parsippany, N.J.

#### 4. Radiophotoluminescence (Glass) Dosimeters

Glass (RPL) dosimeters have, as we believe undeservedly, been somewhat neglected in some countries despite their good reproducibility and stability. The surface of glass dosimeters, in particular those containing much  $\text{Li}_2\text{PO}_3$  or  $\text{B}_2\text{O}_3$ , may soften and become sticky and milky when kept for extended periods at high humidity without proper protection. Simple washing in water, followed by the usual pre-readout cleaning with a detergent, alcohol and/or acetone does, however, completely remove this sticky layer without affecting the stored information or other glass properties. As the glasses are usually encapsulated for use anyway to compensate for their photon energy dependence, for mechanical protection, and in order to exclude intense UV light which either induces or anneals RPL (for reviews, see ref. 1 and 39-41), an increase of their humidity resistance is only of interest in some cases such as *in-vivo* dosimetry. This can be accomplished by modifications in the glass base (42, 43), or by coating the surface with protective layers such as  $\text{SiO}_2$  or non-fluorescent polymers.

More important for practical considerations are a fast build-up and slow fading of the RPL. Unfortunately, both requirements are difficult to meet with the same glass because those with a fast RPL build-up usually also exhibit pronounced fading and vice versa. Build-up and fading kinetics have been studied in great detail as a function of glass base composition, silver concentration, storage temperature, and LET (1, 39, 40). If a glass has a very slow build-up rate such as come glasses with a low silver content (44), or if it has to be read a short

time after exposure, a short stabilizing heat treatment (the optimum temperature treatment depends on the RPL kinetics in the glass to be treated, but for most commercial glasses 15 min. at 100-150°C serves this purpose - Fig. 20) helps to establish the maximum RPL intensity.

Without stabilization, glasses exhibiting RPL which is constant within  $\pm 10$  to 15 % between a few hours and several years after exposure should be considered sufficiently stable at least for personnel monitoring (although RPL glasses have also been suggested for environmental dosimetry, their sensitivity makes them clearly inferior for low dose measurements to some TLD phosphors). Doses of less than 20-50 mrad are difficult to measure with good precision even with the best modern glasses. High precision at higher dose levels (Fig. 21), and unlimited repeatability of measurements makes them particularly attractive for intercomparison studies (45). At least one glass, the FD-2 made by Toshiba, Tokyo, Japan, exhibited less than  $\pm 10$  % deviations between one hour and ten years after exposure during storage at  $22 \pm 2^\circ\text{C}$  (Fig. 22), while others such as the old Bausch & Lomb "High-Z" glass (not now in production) exhibited  $\sim 30$  % fading during the first year.

It has been shown in detailed kinetic studies (39, 40) that increased storage temperature accelerates both build-up and fading. Even at  $30^\circ\text{C}$  and 90 % relative humidity, however, the readings between one day and three months after exposure for three modern RPL glasses (Fig. 23) deviate less than  $\pm 8$  % from the average. If one would accept a pre-readout thermal stabilization, glasses could easily be prepared which are even more fading-resistant than these.

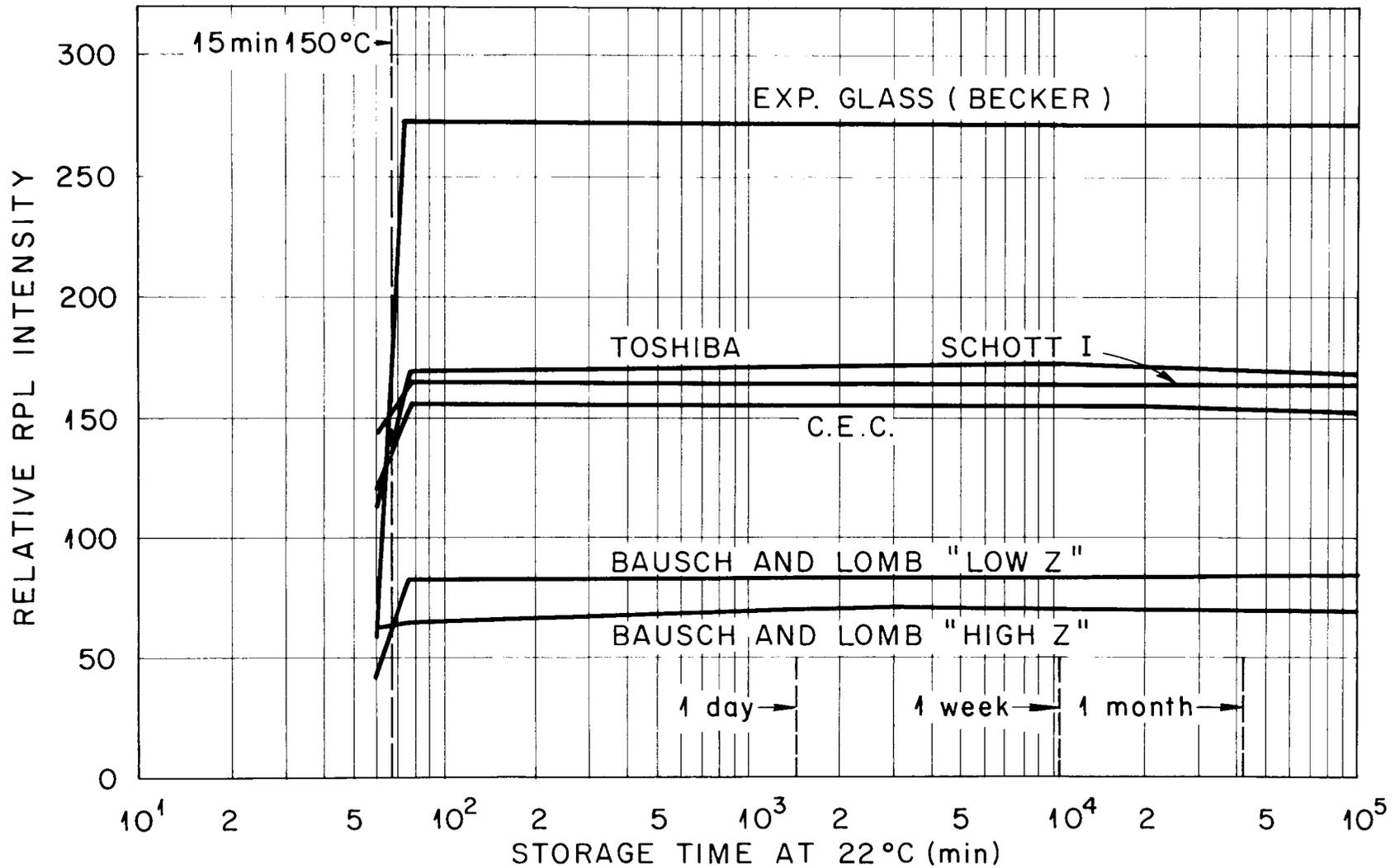


Fig. 20 Storage stability of different RPL glasses of identical dimensions after exposure to the same gamma radiation dose, before and after a stabilizing heat treatment.

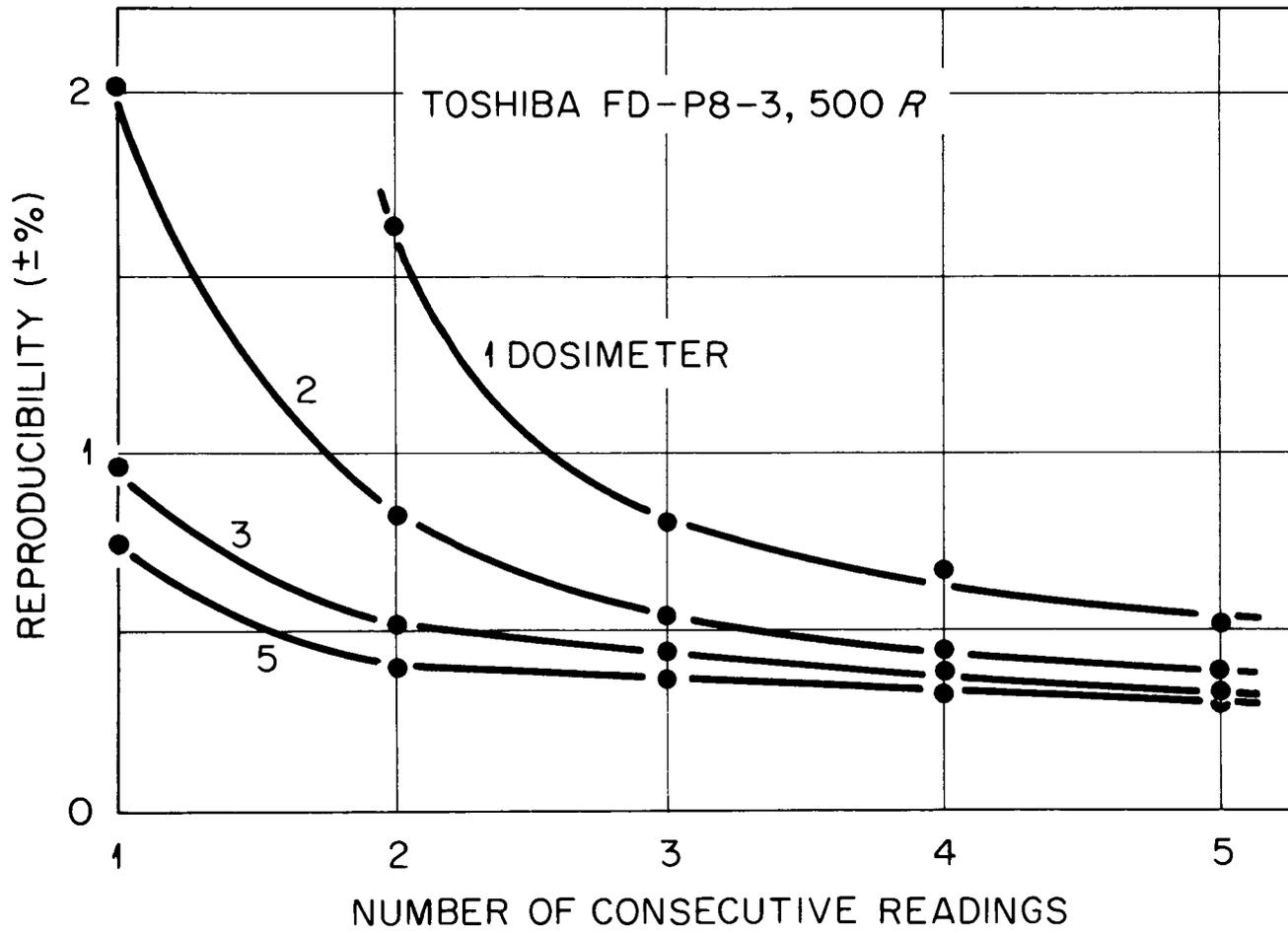


Fig. 21 Accuracy of a high-level dose measurements with a Toshiba RPL glass, for single and multiple read-out of one or more detectors.

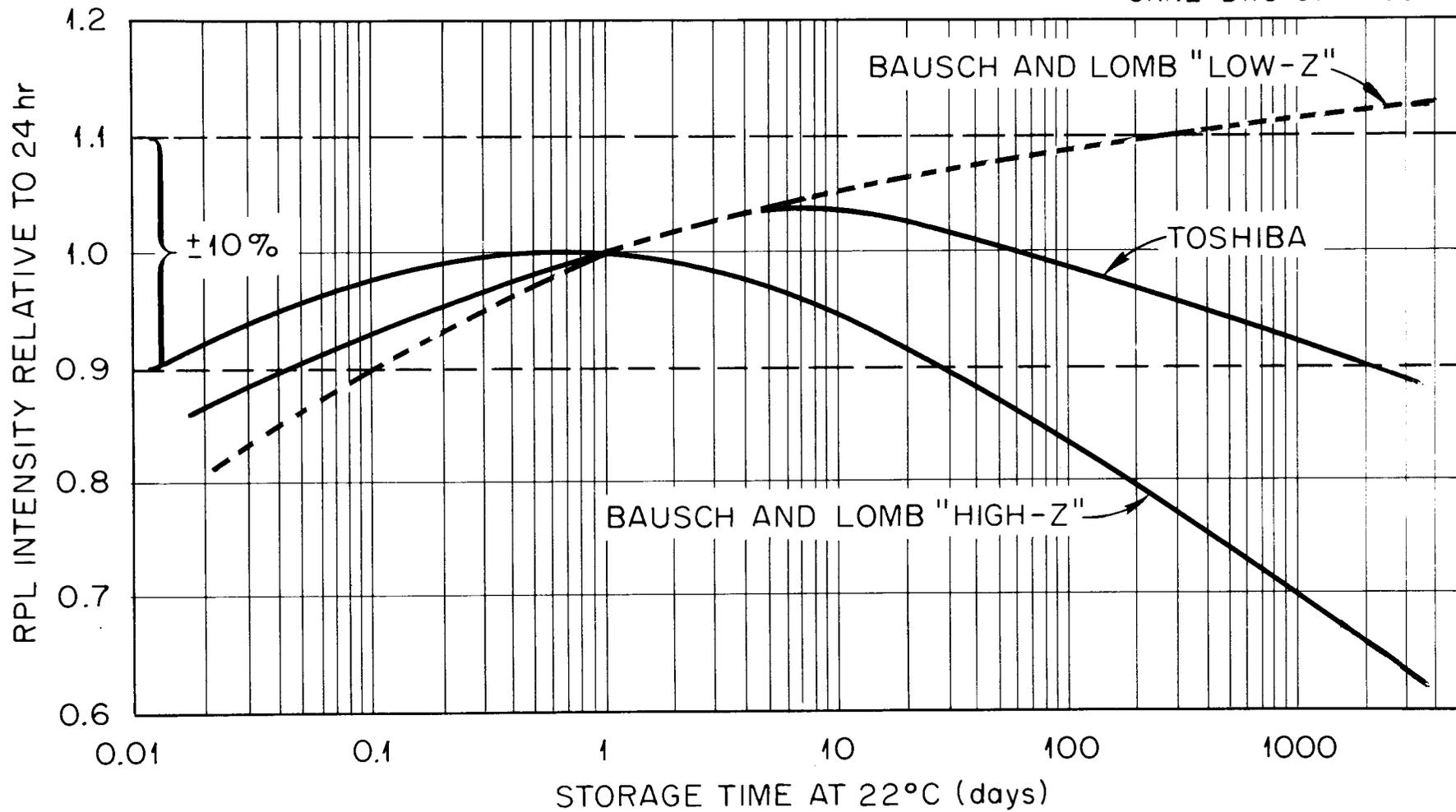


Fig. 22 Build-up and fading of RPL in three types of irradiated dosimeter glasses during storage at  $\sim 22^\circ\text{C}$  in the dark, normalized for the RPL intensity one day after exposure (after 46, 47, and J.S. Cheka, unpublished).

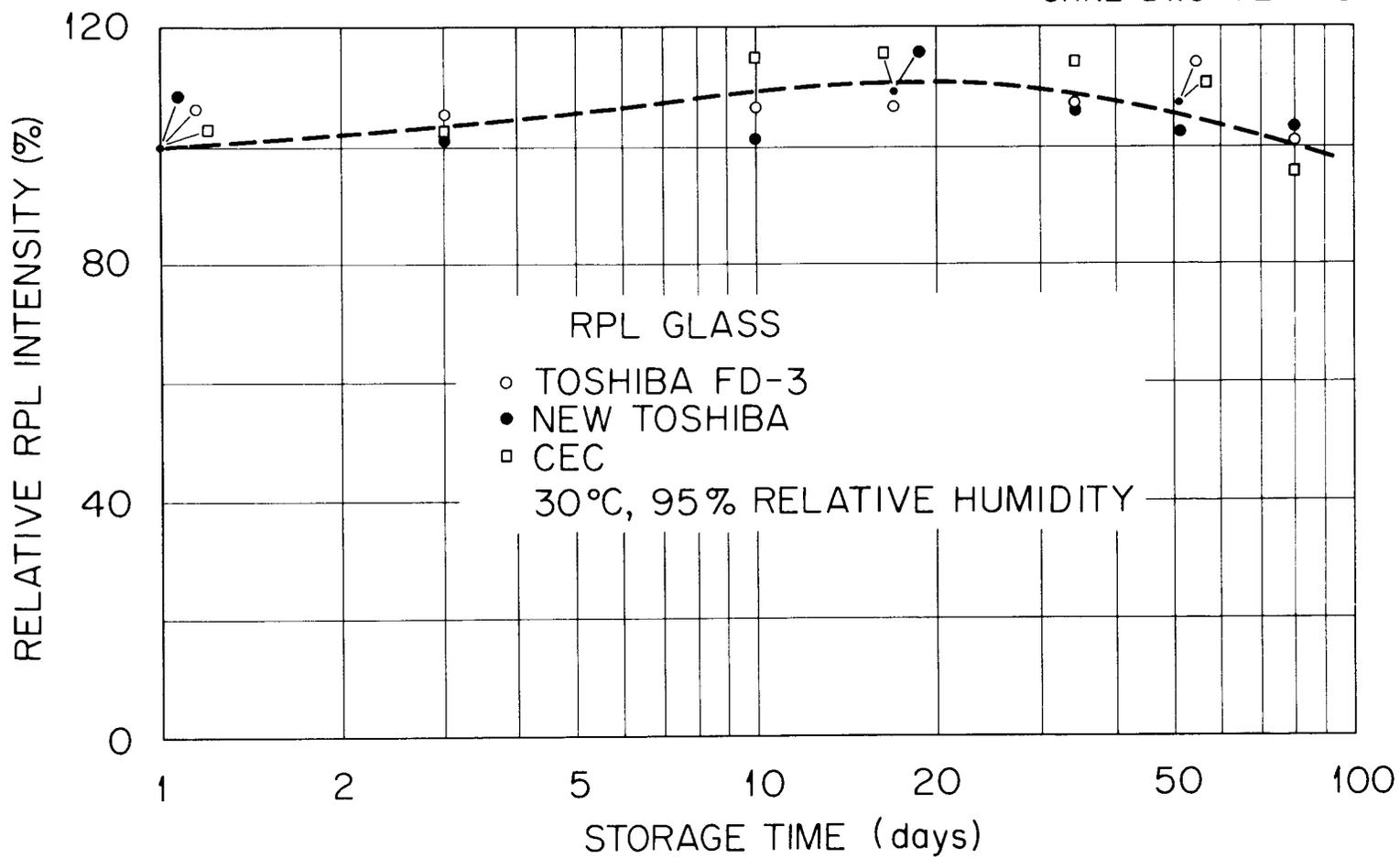


Fig. 23 RPL in three modern dosimeter glasses as a function of storage time at 30°C and 95 % relative humidity, normalized for the response one day after exposure.

## 5. Track Etching Detectors

The use of track etching detectors became a serious alternative to the obviously insufficient nuclear track emulsion for fast neutron dosimetry during the past 5-8 years (for reviews, see ref. 1 and 48). Besides in neutron personnel dosimetry and area monitoring around reactors, accelerators, etc., their use in postal intercomparison of medical and technical neutron sources is currently under discussion.

Between exposure to alpha, recoil particles or (most frequently) fission fragments and the etching of the detectors, the "latent" damaged region along the track can be partially or completely annealed by storage at elevated temperatures without permanent changes in the recording capabilities of the detector. In some sensitive detectors such as cellulose, however, extended storage of the detectors at temperatures above 40-50°C may result in permanent changes of their structure and recording properties (Fig. 24). If the melting point of the detector is as high as it is in some minerals and inorganic glasses, storage at temperatures up to several hundred degrees does not cause any fading even over extended periods of time. In natural crystalline quartz, for example, only 20 % of latent fission fragment tracks fade during 150 days at 710°C (50), and it can be calculated by assuming a simple relationship between fading rate and temperature as illustrated in Fig. 25 that latent tracks in some of these high-melting materials would be stable at room temperature much longer than the age of the solar system (51). Quartz, mica or high-melting glasses have indeed been shown to be usable for track detection at tempera-

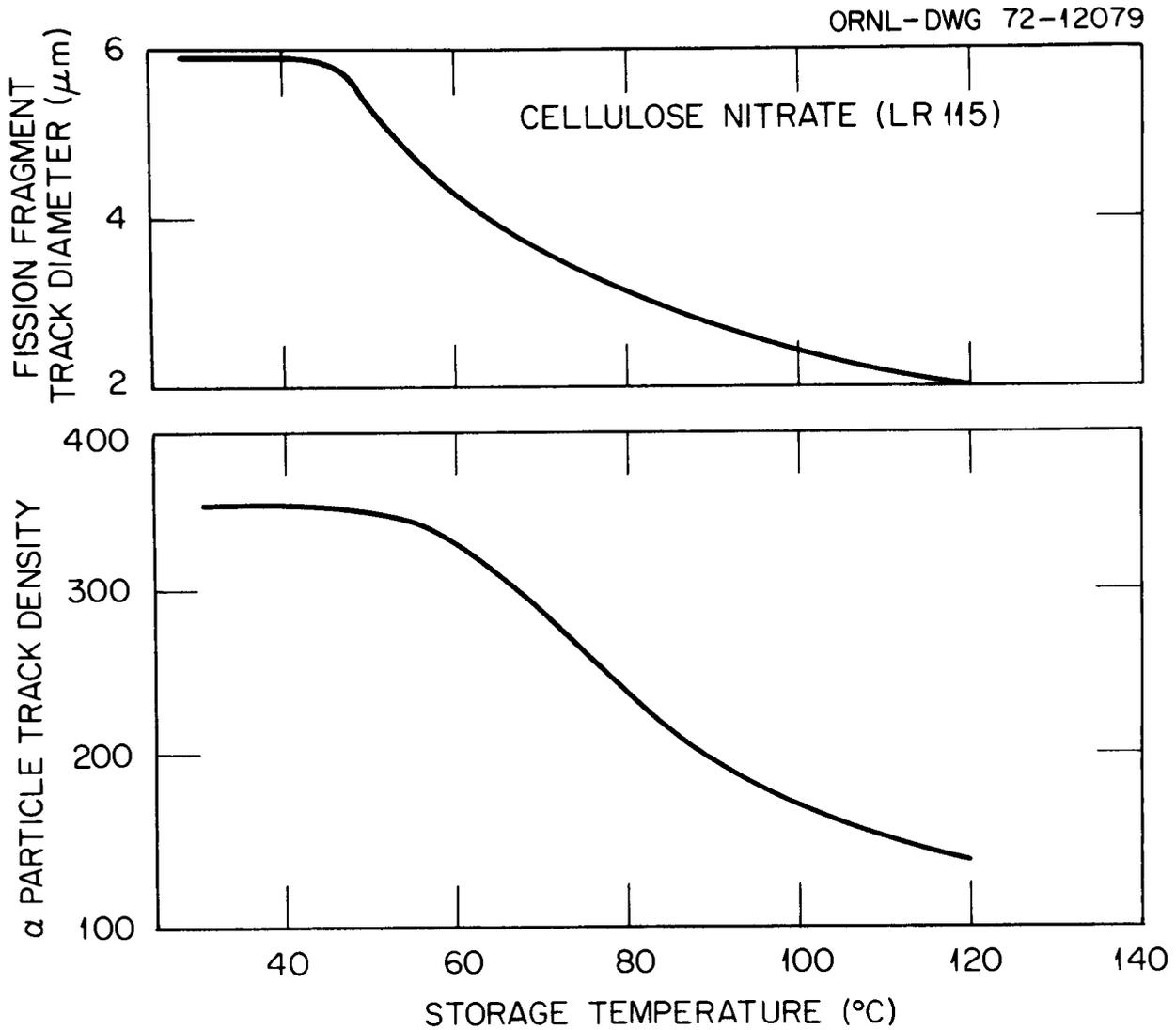


Fig. 24 Effect of extended (4-15 hours) preheating on (a) the diameter of fission fragment tracks; and (b) the relative alpha particle density which is observed in LR 115 cellulose nitrate (Kodak-Pathé, France), irradiated and uniformly etched after the thermal treatment (after 49).

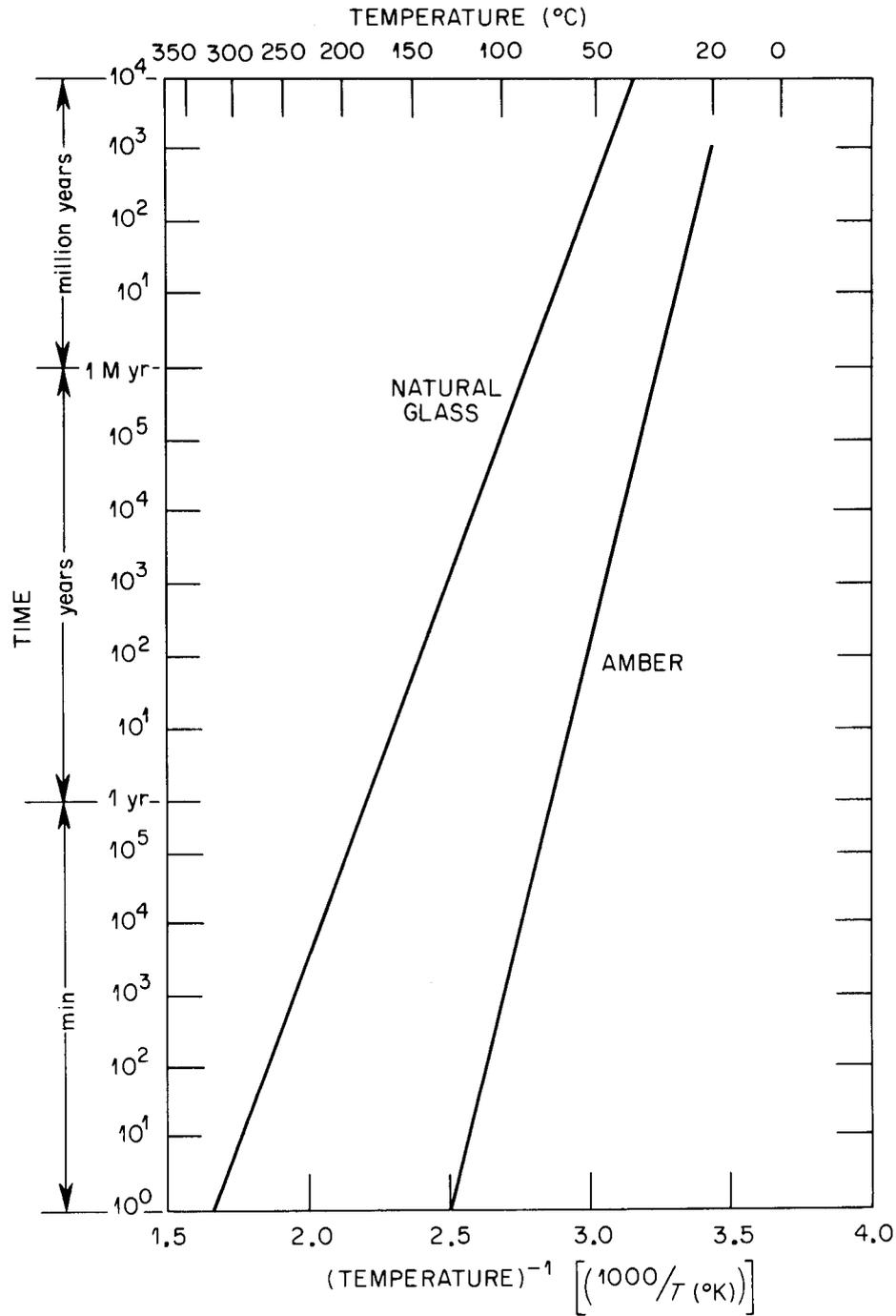


Fig. 25 Latent fission fragment track stability in a basaltic glass from the Mid-Atlantic Ridge (after 83), and in amber (after 84), as determined by measuring, at various temperatures, the time required for total annealing of the latent tracks.

tures exceeding several hundred °C, for example in smoke stacks or inside a reactor. Some conventional glasses, however, such as microscope slides, exhibit substantial fading already after storage for several days at 60 to 100°C (52).

In polymers, no fading of the latent tracks has been observed at temperatures below about 50°C. Even in 10 µm polycarbonate foils stored for three months at 30° and 95 % relative humidity before etching and spark-counting of the fission fragment tracks, no fading could be detected (Fig. 26). Only if the softening temperature of a plastic is approached does rapid annealing begin to take place. For example, the accumulated "background" alpha radiation effect due to atmospheric radon, etc. in a cellulose nitrate foil can be annealed prior to use by overnight storage at ~ 80°C (85). In a commercial 10 µm cellulose nitrate foil (LR 115 made by Kodak-Pathé, France), pronounced fading of latent alpha particle tracks has been induced by one hour of storage at 70°C. It has been suggested that fading during the etching at the usual temperature of 60°C competes in this material with the process of track amplification, and the etching temperature should consequently be lowered (65).

In other foils such as polycarbonate (Makrofol), cellulose triacetate (Triafol T), or cellulose butyroacetate (Triafol B - all made by Bayer, Leverkusen, Germany), higher temperatures are required for annealing (53, 54). As can be seen in Fig. 27 and 28, fission fragment tracks in Makrofol E and Triafol B are still quite stable at 60°C, but the fading rate is rapidly accelerating around 100°C. Extended etching can make

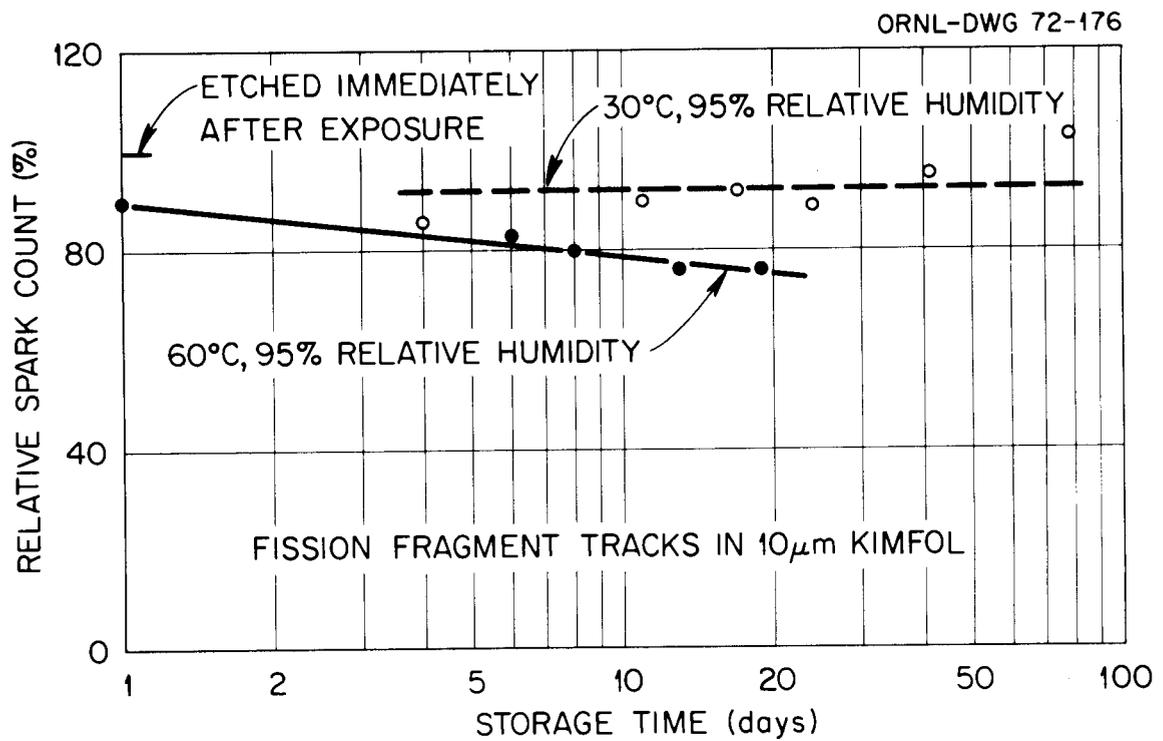


Fig. 26 Relative number of fission fragment spark counts in 10  $\mu$ m polycarbonate (Kimfol) as a function of post-exposure, pre-etching storage time of the foil under adverse climate conditions.

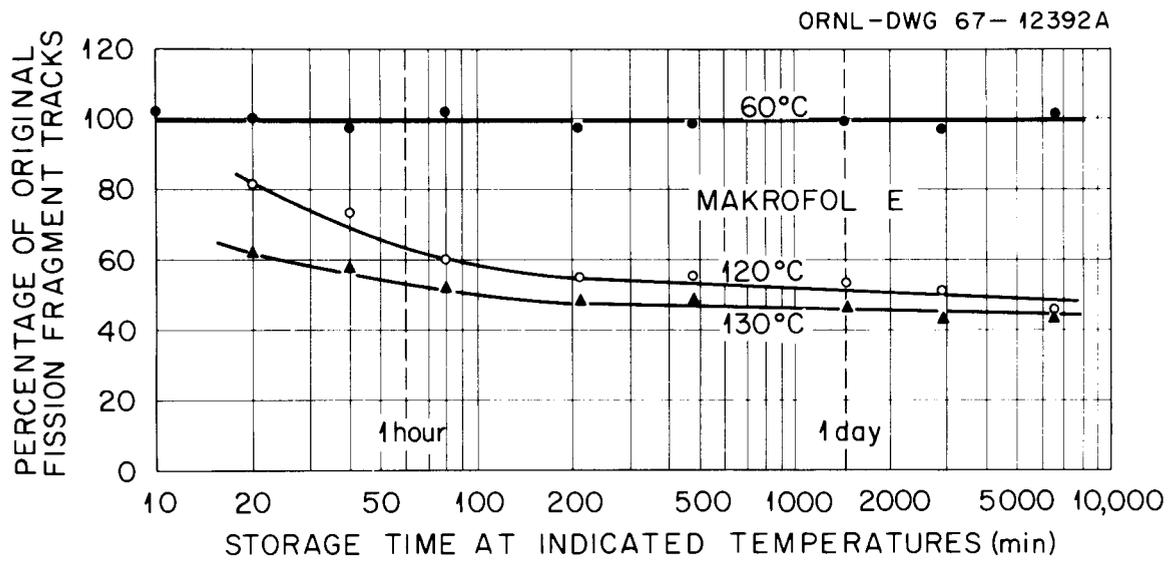


Fig. 27 Percentage of original fission fragment tracks in polycarbonate (Makrofol E), and in cellulose acetobutyrate (Triafol B) which remain visible after storage of the foil prior to etching at various temperatures.

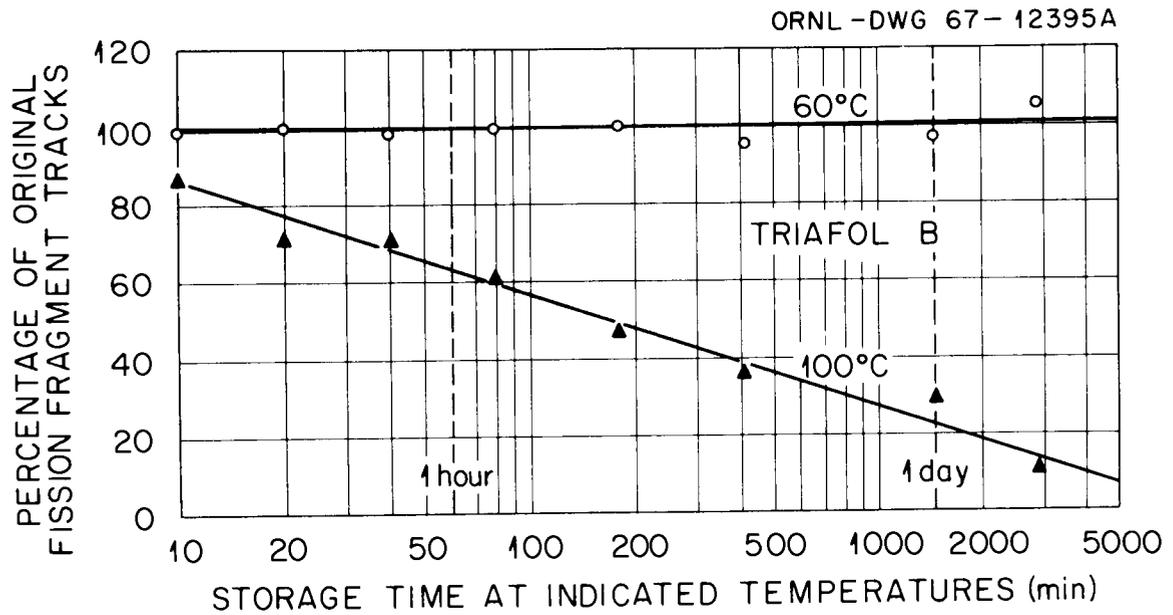


Fig. 28 Percentage of original fission fragment tracks in polycarbonate (Makrofol E), and in cellulose acetobutyrate (Triafol B) which remain visible after storage of the foil prior to etching at various temperatures.

some "invisible" tracks reappear. Of course, not only the number of visible etch pits or tracks, but also their dimensions (size distribution, length) undergo pronounced changes during fading; such changes may be used as an indicator of the detector's thermal history (55, 56).

The fading kinetics in polymers are affected by several parameters other than temperature. For instance, exposure of a cellulose nitrate foil to intense ultraviolet light induced fading of latent alpha particle tracks (57). High-LET tracks, such as those of fission fragments, tend to fade less rapidly than alpha-particle tracks in a given material (this process can to some extent be used for discrimination against low-LET particle tracks). The atmosphere in which a foil is stored between exposure and etching also may have a strong effect on the etching parameters. As can be seen in Fig. 29, little change is observed in Lexan during storage in air and inert gases, but the track length substantially changes in oxygen and in vacuum. There is also evidence that the fading in some polymers is slightly accelerated at high relative humidities, and that "old" tracks behave differently from "young" tracks in minerals and glasses. Apparently, physical as well as chemical processes contribute to the thermal fading of latent tracks. Although polymer track detectors are already widely used, no field tests of their fading stability have yet been carried out.

#### 6. Miscellaneous Solid-State Detectors

Wide-based  $n^+pp^+$  junction type silicon diodes, whose forward resistance is semi-permanently changed as a function of neutron exposure, have been suggested as high dose-level (> 5-10 rad) fast neutron

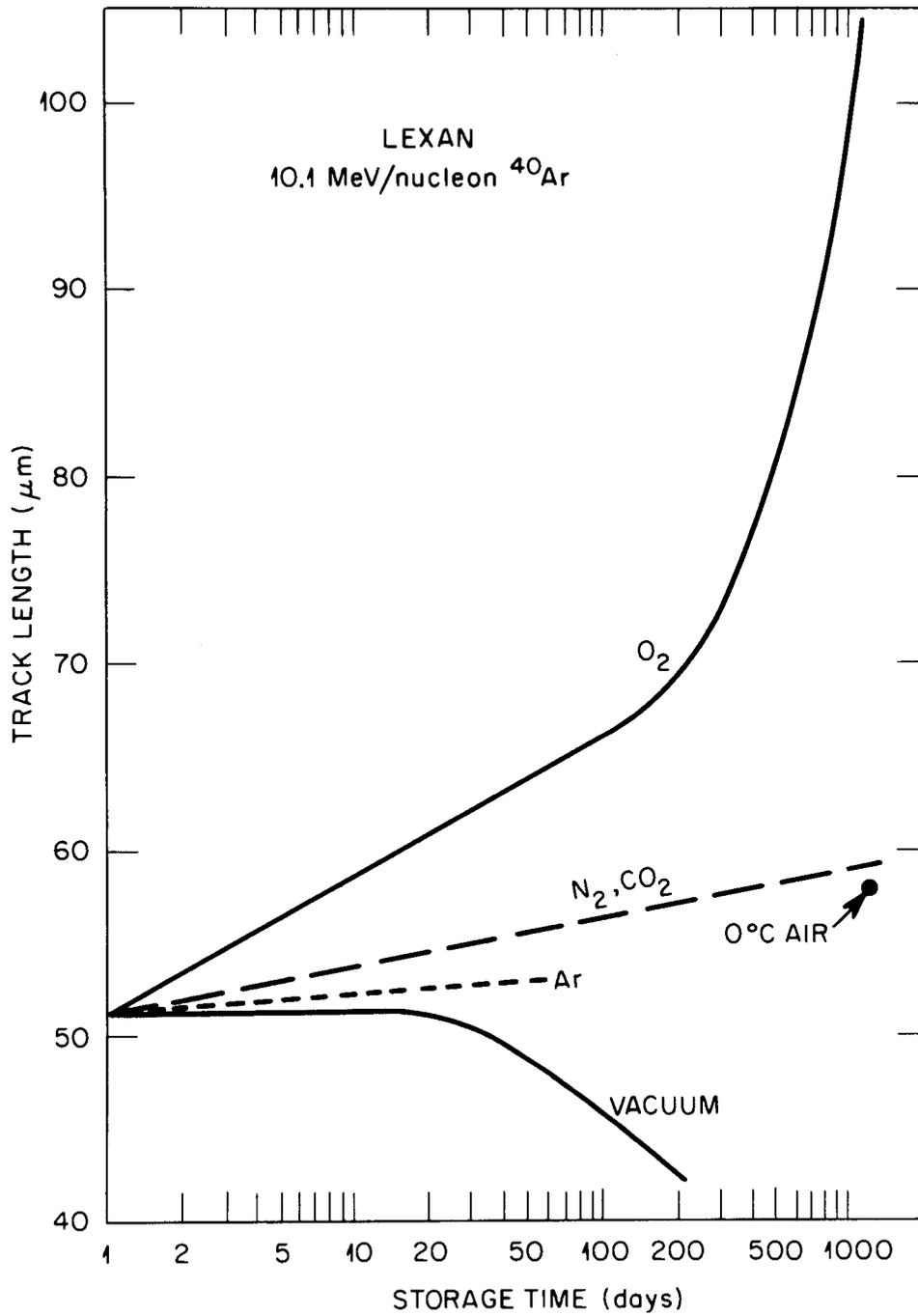


Fig. 29 Effect of the gas in which Lexan has been stored in the dark at room temperature, between exposure to 10.1 MeV/nucleon  $^{40}\text{Ar}$  ions and etching, on the observed track length (after 87).

dosimeters (for a review, see ref. 1). Reports by various investigators on the stability of the neutron-induced effect in different types of such diodes vary: For the U.S. diodes, 8 % fading was found between 1 min. and 30 min. after exposure (58), while another author (59) reports only 3.5 % during one day, and 26 % fading after 100 days of storage at room temperature. Under similar conditions, 15 % fading has been reported after 100 days in U.K. diodes, and 18 % in Swedish diodes (60, 61), while others (62) report 20 to 23 % in Swedish as well as German diodes. Perhaps the disagreements are largely due to inconsistencies in the irradiation time, storage temperature, and the exact time at which the first post-irradiation measurement is taken (the initial fading during the first  $\sim$  30 min. is quite rapid (58)). The fading rate tends to increase slightly with dose level.

The fading proceeds more rapidly at increased temperature, and a short temperature treatment of the irradiated diodes after exposure (2 minutes at 100°C are suggested (62)) helps to complete the initial fading, thus "stabilizing" the radiation effect in the diodes. No field stability tests have yet been carried out with these diodes.

Dosimeters based on thermally stimulated exoelectron emission (TSEE) and, to a lesser degree, optically stimulated EE, are the most recent addition to the arsenal of sensitive integrating solid-state dosimeters (for recent reviews of this rapidly expanding field, see ref. 1, 63). The physical process of electron leakage from populated traps is apparently governed by the same laws as in TLD, shallower traps (low-temperature peaks) being subject to more rapid fading than

on a graphite disk during one month at 60°C, and even at 150°C only little fading occurred during the first few hours, as one would expect for a peak temperature of  $\sim 270^\circ\text{C}$ . Later, extensive studies at ORNL with ceramic BeO disks (Thermalox 995 manufactured by Brush Beryllium Co., Elmore, Ohio), revealed, however, that even a stabilization treatment consisting of extended heating of the detectors at 1450°C, followed by saturation of the surface with water (69) could not always prevent slow changes in the material's TSEE characteristics, which manifest themselves as fading and/or fluctuations in the detector sensitivity which are frequently rather erratic.

Although some progress has been made in understanding these changes, and some well-equilibrated ceramic BeO samples have been employed at ORNL in personnel and environmental monitoring field tests with no detectable fading or change in sensitivity for periods up to several weeks (70), other experiments, in particular at elevated temperatures and humidities, still show more or less pronounced changes. These effects are presently being studied at ORNL. Permanent encapsulation of the exoelectron emitter would, of course, avoid the disturbing effect of atmospheric constituents; light, which is known to induce as well as anneal signals (depending on the emitter, wavelength, and pre-exposure level); mechanically induced tribo-signals; and surface contamination. Indeed such systems have been made, and are evidently being used in the Soviet Union (71).

There are numerous other radiation effects in solids which have been used or considered for radiation dosimetry. The build-up and, more frequently, the fading of coloration or discoloration in glasses and

deep traps. In  $\text{CaSO}_4$ , for example, a lower-temperature peak at  $170^\circ\text{C}$  disappeared completely after 5 days at  $60^\circ\text{C}$ , while another peak at  $\sim 280^\circ\text{C}$  remained completely unaffected by such a temperature treatment. Even apparently single-peak materials such as  $\text{BeO}$  exhibit a shift in the peak temperature during fading at  $150\text{--}180^\circ\text{C}$  indicating the presence of a trap depth distribution, the shallower ones being annealed first. Apparently, no retrapping takes place during fading (for a summary of earlier TSEE fading studies, see ref. 64).

In some TSEE emitters such as  $\text{LiF}$ , the fading rate was found to be independent of humidity, and the same in air and vacuum (64). In the majority of cases, however, secondary surface processes complicate the fading kinetics. As exoelectron emission is likely to occur only from an extremely thin ( $< 100 \text{ \AA}$ ) surface layer and the active TSEE centers in some substances are known to consist of adsorbed or chemisorbed oxygen, the strong effect of gas or humidity adsorption and desorption processes on the emission characteristics is easily explainable, and has been the subject of numerous investigations (see, for example, 66, 67). For example, a  $\text{BaSO}_4$  detector exhibited 5 % fading during 5 days at  $\sim 22^\circ\text{C}$  if kept in a sealed tube, but 60 % if the material was plated on an open carrier (Fig. 30). In  $\text{CaSO}_4$ , fading is substantially accelerated at high ambient humidity, probably due to the addition of crystal water. The fading kinetics in  $\text{BeO}$  are also different from those measured in air if kept in water vapor or vacuum.

In  $\text{BeO}$ , which is one of the more attractive TSEE materials due to its low-Z, high sensitivity, and convenient peak location, no fading was observed in early experiments (68) with powdered materials plated

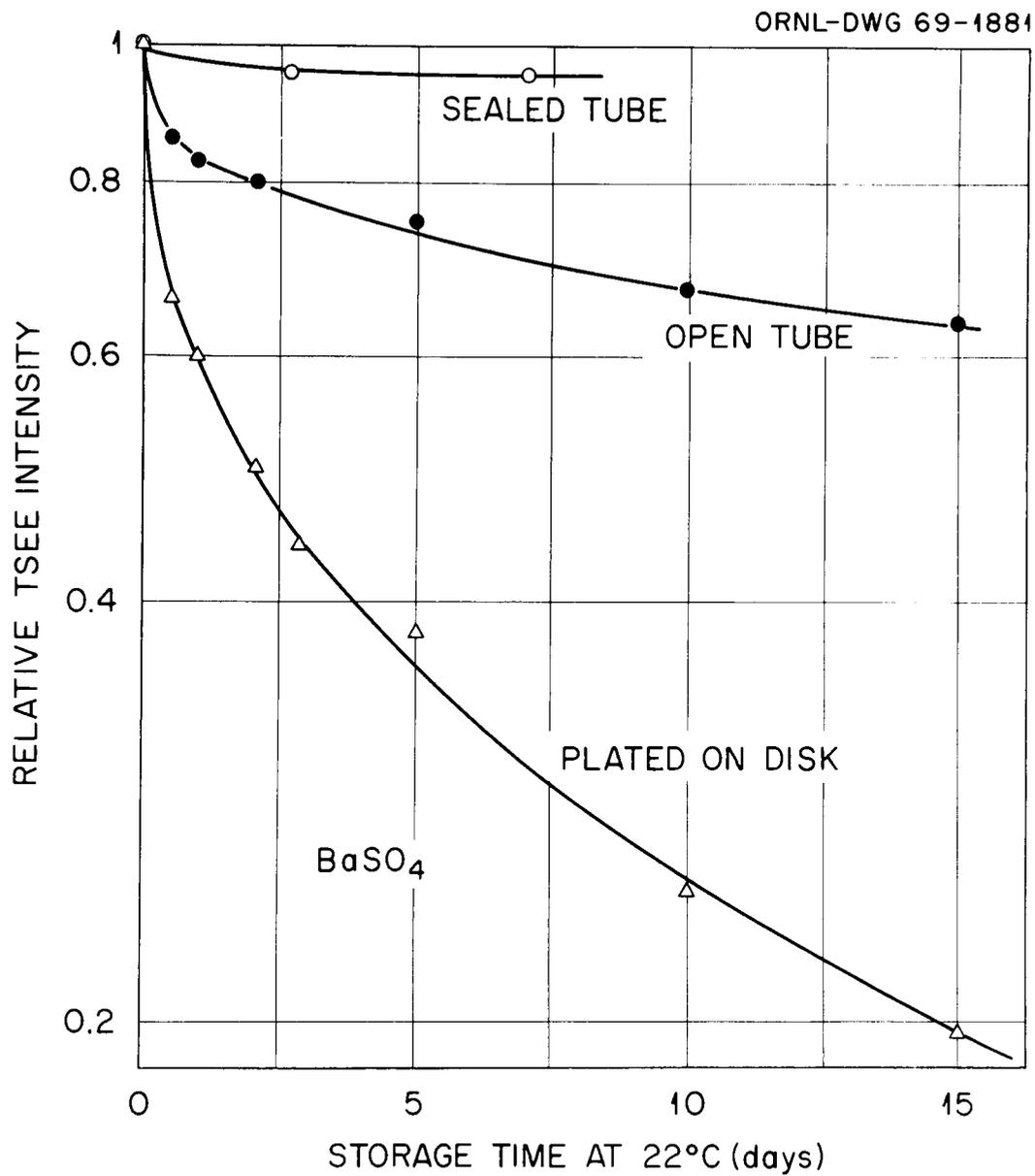


Fig. 30 Fading of TSEE in  $\text{BaSO}_4$  at room temperature if kept in a sealed tube, an open tube, and plated on the surface of an unprotected flat disk (after 86).

polymers is known to depend strongly on storage temperature, in other cases such as fluorescence degradation also on the ambient level of oxygen (for a review, see ref. 1). As these detectors for high dose level measurements are usually evaluated shortly after irradiation, their long-term fading stability is normally only of minor interest. Also not to be discussed here are numerous more exotic techniques which have not yet reached the stage of practical application.

### 7. Conclusions, and Desirable Follow-up Studies

It is obvious that the currently still wide-spread and in some countries rapidly expanding use of photographic film in personnel dosimetry should be discouraged in areas which have during all or part of the year a humid, warm climate. Although this fact has been self-evident to many experts in this field for years, educational efforts are still required as long as many international and national organizations, as well as individual radiation protection experts and administrators, strongly support the use of film even in exceedingly hot and humid countries such as Brazil and India (see, for example, ref. 72). Three tolerance levels of the film can easily be distinguished:

1. Nuclear track emulsions for fast neutron measurements should not be used without additional sealing of each individual film under any circumstances with the possible exception of short-term (less than one week) low-dose, high neutron energy measurements in dry, cool desert climates. Careful sealing of the dessicated film in a plastic-aluminum compound foil somewhat reduces the fading rate, but even so its use for periods of more than a few days in a humid and warm

place is not recommended.

2. Although the use of X- and gamma radiation monitoring films is not restricted by fading and fogging for periods up to several months in a cool or moderate climate and/or air-conditioned facilities, they should not be employed without additional careful sealing after dessication in tropical countries, where rapid fading takes place and leads to a dangerously misleading underestimation of the actual radiation exposure. In addition to the lack of stability, there are, of course, numerous other disadvantages of the film dosimeters. All performance comparisons which have been reported between film and TLD dosimeters show indeed that the film dosimeters are less accurate (ref. 73-78, to quote only a few), and TLD is replacing film in the more advanced institutions (79).
3. Careful sealing of dessicated X +  $\gamma$  films in metal-plastic compounds increases their fading resistance, but there are still areas which have such a severe climate that this protection is insufficient.

It has been suggested to reduce the possible errors due to film fading by storing films which have been exposed to a known dose at each location (institute, city, hospital, etc.) where films are worn. This method may indeed be a good indicator for excessive fading, but its practical value is rather limited not only because of the additional labor and costs, but also because

1. no fading "corrections" are practical whenever total or almost total fading takes place;
2. it is frequently not known whether a given personnel exposure occurred at the beginning or towards the end of a monitoring period; and

3. the climatic exposure of individual badges may differ substantially from the "average" at a given location.

Additional tests would be desirable, mostly with photographic films, in order to exactly establish the limitations in their usability:

1. As different emulsion types behave differently, more types of dosimeter films should be included in such tests;
2. the performance of the sealing of the Kodak-Pathé Type 1 film should be investigated (the actual Type 1 emulsions which have to be read by inaccurate reflection densitometry have, however, to be replaced by normal dosimeter films).
3. different films should be sealed, after careful dessication, with and without additional dessicants\*) in each individual package; an additional testing of the film sealings for possible leaks could be performed by placing them in vacuum;
4. the fading rates in films exposed to X-rays and gamma radiation should be compared (there may be less fading in the films exposed to lower photon energies);
5. exposure to higher and lower gamma dose levels should permit the more accurate evaluation of the fading in some emulsions;
6. instead of having just one storage time of several months, the effect of different shorter storage times (such as 1, 2, 4, and 8, weeks) should also be studied; and

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\*)Molecular-sieve type Zeolites appear to be most attractive for this purpose, because  $\text{CaCl}_2$  could affect the film.

7. several additional locations for dosimeter storage should be selected, preferably with a more detailed climatic record (exact times during which certain temperatures and humidities have been exceeded).

Many of these follow-up experiments could be carried out by local scientists in the countries concerned, but some orientation, coordination and advice will be required in order to guarantee reliable, comparable results.

It is, of course, fully realized that the question of detector stability is only one of many aspects to be considered in the choice of an optimum dosimetry system. Other factors such as costs; reliability; dose and energy range; weight; mechanical stability; speed, convenience and accuracy of readout; legal and psychological considerations; training level of local staff; etc., may be equally or even more important (for a more detailed discussion, see ref. 1 and 2). Nevertheless, the role of stability problems in countries with an at least partial tropical climate should, as this report attempts to demonstrate, not be underestimated.

Some TLD phosphors are far superior to the film, in particular  $\text{CaSO}_4:\text{Dy}$ , which is eminently suitable for environmental monitoring. The fading in  $\text{LiF}:\text{Mg,Ti}$  should not be neglected, but appears to be tolerable for personnel monitoring, perhaps even for intercomparison studies if a build-in "fading-check" is provided. Some other promising phosphors such as  $\text{CaSO}_4:\text{Tm}$ ,  $\text{BeO}$ , and  $\text{Mg}_2\text{SiO}_4:\text{Tb}$  should be included in further field testing. Laboratory tests with RPL glass dosimeters and some track etching detectors indicate sufficient stability at warm climates, but fading tests under field conditions with both systems may

be desirable. The long-term use of silicon diodes under tropical conditions is questionable, and more work on the stability of TSEE dosimeters is required.

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Figure Captions

- Fig. 1 Developed Kodak Personal Monitoring Film Type 2, heavily damaged during several weeks of storage at 30°C and 95 % relative humidity.
- Fig. 2 Thermal fogging of an unirradiated dosimeter film during storage for various times at different elevated temperatures (after 5).
- Fig. 3 Fading and fogging in the insensitive emulsion of a Kodak Personal Monitoring Film Type 2 during three months of storage in a simulated tropical climate at 30°C, 95 % relative humidity.
- Fig. 4 Latent image fading in a Kodak RM dosimeter film at 30°C and different relative humidities (after 32).
- Fig. 5 Optical density of a gamma-irradiated Kodak Personal Monitoring Film Type 2 (sensitive emulsion) as a function of storage time between exposure and (simultaneous) processing of the films; storage in standard laboratory atmosphere, and in a protected space outdoors with or without additional sealing in a thin polyethylene bag, in a subtropical climate. Note the wide scattering of the O.D.s in the stored films.
- Fig. 6 Fading of the total optical density in a Kodak PM Type 2 film after exposure to 400 mR of gamma radiation during storage at different climates (after 6).
- Fig. 7 Visible recoil proton track density in Kodak NTA film as a function of storage time at 20°C, and 75 % and 90 % relative humidity, if the film (without additional wrapping) is irradiated after careful drying; and at 75 % relative humidity,

if permitted to equilibrate prior to neutron irradiation.

Fig. 8 Relative visible recoil proton track density in Kodak NTA film exposed to Po/Be neutrons as a function of storage time at different humidities.

Fig. 9 Relative track density in Kodak NTA film for storage at different temperatures and humidities (after 29).

Fig. 10 Fading of latent image in a Kodak NTA film in a semi-tropical climate.

Fig. 11 Water content of the gelatin in a nuclear track emulsion at 20°C as a function of relative air humidity (after 33).

Fig. 12 Track fading in Kodak NTA film at ambient temperature and 100 % relative humidity in normal wrapping; placed in a protective thick polyethylene holder; and with both film and holder carefully dried before sealing (after 28).

Fig. 13 Kodak PM Type 2 films uniformly irradiated at the same time to 0.4 R gamma radiation and

- a. stored for three months in Bogota, Colombia;
- b. unsealed in Cartagena, Colombia;
- c. sealed in Cartagena;
- d. unsealed in Guayaquil, Ecuador; and
- e. sealed in Guayaquil.

On the lower right side, unexposed control film.

Fig. 14 Simplified diagram of the effect of storage temperature on the fading rates in three sensitive TL phosphors.

- Fig. 15 Fading and build-up of the TL signal in  $\text{Mg}_2\text{SiO}_4:\text{Tb}$  at different storage temperatures (after 80).
- Fig. 16 Fading of TL in  $\text{CaSO}_4:\text{Mn}$  at 25 and 37°C (after 81), and at 26°C (after 82).
- Fig. 17 Fading of TL in  $\text{CaF}_2:\text{Dy}$  at different storage temperatures.
- Fig. 18 Fading of TL in  $\text{LiF}:\text{Mg,Ti}$  (TLD-100) powder at different storage temperatures.
- Fig. 19 Fading of TL in  $\text{CaSO}_4:\text{Dy}$  powder at different storage temperatures.
- Fig. 20 Storage stability of different RPL glasses of identical dimensions after exposure to the same gamma radiation dose, before and after a stabilizing heat treatment.
- Fig. 21 Accuracy of a high-level dose measurements with a Toshiba RPL glass, for single and multiple read-out of one or more detectors.
- Fig. 22 Build-up and fading of RPL in three types of irradiated dosimeter glasses during storage at  $\sim 22^\circ\text{C}$  in the dark, normalized for the RPL intensity one day after exposure (after 46, 47, and J.S. Cheka, unpublished).
- Fig. 23 RPL in three modern dosimeter glasses as a function of storage time at 30°C and 95 % relative humidity, normalized for the response one day after exposure.
- Fig. 24 Effect of extended (4-15 hours) preheating on (a) the diameter of fission fragment tracks; and (b) the relative alpha particle density which is observed in LR 115 cellulose nitrate (Kodak-Pathé, France), irradiated and uniformly etched after the thermal treatment (after 49).

- Fig. 25 Latent fission fragment track stability in a basaltic glass from the Mid-Atlantic Ridge (after 83), and in amber (after 84), as determined by measuring, at various temperatures, the time required for total annealing of the latent tracks.
- Fig. 26 Relative number of fission fragment spark counts in 10  $\mu\text{m}$  polycarbonate (Kimfol) as a function of post-exposure, pre-etching storage time of the foil under adverse climate conditions.
- Fig. 27 and 28 Percentage of original fission fragment tracks in polycarbonate (Makrofol E), and in cellulose acetobutyrate (Triafol B) which remain visible after storage of the foil prior to etching at various temperatures.
- Fig. 29 Effect of the gas in which Lexan has been stored in the dark at room temperature, between exposure to 10.1 MeV/nucleon  $^{40}\text{Ar}$  ions and etching, on the observed track length (after 87).
- Fig. 30 Fading of TSEE in  $\text{BaSO}_4$  at room temperature if kept in a sealed tube, an open tube, and plated on the surface of an unprotected flat disk (after 86).



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