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*High-Energy Neutron Dosimetry
at the Clinton P. Anderson
Meson Physics Facility*

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HIGH ENERGY NEUTRON DOSIMETRY AT THE
CLINTON P. ANDERSON MESON PHYSICS FACILITY

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ABSTRACT

Neutron energy spectrum measurements performed at the Clinton P. Anderson Meson Physics Facility indicated potential areas for high energy neutron exposure to personnel. The low sensitivity of the Los Alamos thermoluminescent dosimeter (TLD) to high energy neutrons warranted issuing a NTA dosimeter in addition to the TLD badge to employees entering these areas. The dosimeter consists of a plastic holder surrounding NTA film that has been desiccated and sealed in a dry nitrogen environment. A study of the fading of latent images in NTA film demonstrated the success of this packaging method to control the phenomenon. The Los Alamos NTA dosimeter is characterized and the fading study discussed.

INTRODUCTION

The Clinton P. Anderson Meson Physics Facility (LAMPF) is one of the largest and most powerful nuclear science facilities in the world.¹ Operated by the Los Alamos National Laboratory of the University of California for the United States Department of Energy, LAMPF is an international research facility available for use by qualified members of the world's scientific community.

The heart of the installation is a half-mile-long linear accelerator (linac) that provides beams of protons with energies up to 800 MeV at an average current of 1 mA. LAMPF generates a proton beam more intense than that from all comparable accelerators in the world combined.

The Los Alamos Laboratory is committed to insuring the health of its employees. In accordance with Department of Energy Order 5480.11, the Radiation Protection Group at Los Alamos is responsible for monitoring radiation exposure to personnel and visitors. The Los Alamos thermoluminescent dosimeter badge (TLD) has provided an accurate and versatile instrument for the measurement of ionizing radiation. Recently, unique experimental environments at LAMPF have required the development of a dosimeter to effectively measure the dose component due to high energy neutrons (> 10 MeV). The sensitivity of the Los Alamos TLD badge decreases sharply with increasing neutron energy. Therefore, employees who work in areas where there is a potential for a significant exposure from these high energy neutrons are provided with an additional dosimeter which uses NTA film. NTA is a nuclear emulsion which is relatively sensitive to neutrons above 1 MeV. The double badging is maintained throughout the operating period of the LAMPF accelerator and its associated experimental areas and facilities.

The primary technical difficulty in using NTA film is the latent image fading of proton tracks in the nuclear emulsion between exposure and processing. Relative humidity and temperature have been shown to dominate the rate of fading. A special packaging method is employed at Los Alamos to reduce the fading effect. The packaging process involves dry nitrogen desiccation of the NTA film and hermetically sealing it in a plastic pouch while in the nitrogen environment.

HISTORY

In 1978, the Los Alamos Scientific Laboratory converted from a photon-NTA combination film badge to a TLD badge. The new TLD badge delivered a more accurate measurement of occupational exposure to radiation while greatly simplifying dosimeter processing. The TLD-600 Harshaw LiF chip present in the dosimeter also provided a sensitivity to low-energy neutrons not previously possible with the NTA film badge alone. A detachable holder for NTA film was included in the TLD badge design to facilitate fast neutron measurement. However, this configuration proved to be unacceptable for two reasons. The detachable holder design does not preclude the fading problem observed for NTA film. Furthermore, the additional holder would invalidate the calibration, and thus the DOE/LAP certification of the TLD badge, which was based on the TLD badge without the NTA holder.

In 1987, neutron energy spectrum measurements were made at a potentially high neutron-energy location at LAMPF.² Neutron survey instruments along with personnel dosimeters were placed in the ER-1 experimental area with the proton beam being stopped in a carbon block in an adjacent beam line. The experiment was performed to evaluate the neutron energy spectrum and dose equivalent rate under conditions of controlled beam spill and to assess the relative sensitivity of each neutron measuring device. Computer codes unfolded data from various detectors which included moderated, unmoderated, and cadmium covered TLD 600 and TLD 700 LiF chips. In addition, a plastic scintillator threshold activation detector, C(n,2n), and a bismuth fission chamber were used. The unfolding codes revealed that more than 90% of the neutron dose equivalent was due to neutrons of energy greater than 1 MeV, and 70% due to neutrons of energy greater than 10 MeV.

The experiment further showed that the Los Alamos TLD badge underresponded by a factor of 5 to 7 for this particular neutron spectrum.

NTA film similarly underresponded but only by about 20%. However, the sum of the two dosimeters was in good agreement with the spectrum unfolding results, as would be anticipated based on their sensitivity ranges.

In 1988, employees at LAMPF who entered ER-1 were issued a separate NTA badge in addition to the regular TLD badge. The NTA film was placed inside a cadmium covered Cycolac plastic TLD badge holder. The double badging system continued throughout LAMPF's 1988 operating period. High relative humidities from extensive rainfall during this period prompted a concern for fading.

A fading study was subsequently initiated to recharacterize the NTA dosimeter. Badges irradiated to 500 mrem (PuBe-238 or Cf-252) were developed throughout a 2-month period. More than 50% fading was observed in the first 20 days since irradiation and no tracks were present at 60 days. The literature was reviewed in order to understand the fading phenomenon in NTA film and to determine a possible solution for future LAMPF operating periods.

LATENT IMAGE FORMATION AND FADING

Kodak personal neutron track monitoring film, Type A (NTA), features sensitive material of silver halide crystals dispersed in gelatin.³ A thin layer of this emulsion is coated on one side of a cellulose acetate base. Although neutrons themselves do not expose the film, elastic scattering

reactions between the incident neutrons and hydrogenous nuclei in the emulsion, the film base, the film packaging, or the plastic film holder generate recoil protons. These recoil protons leave tracks in the film which may be developed later. A measure of exposure can then be inferred from the track population.

Latent image formation occurs in two steps. Electrons of the bromide ions are raised to vacant states in the conductance band of the crystal by the passage of the recoil proton. These electrons migrate through the crystal until they are trapped at sites on the surface called sensitivity specks. These locations are actually silver clumps or impurity atoms. Meanwhile, the excess bromide ions slowly diffuse out of the crystal. The now negatively charged sensitivity specks attract silver ions moving freely within the lattice and combine to form silver atoms. A sufficient number of silver atoms on the crystal surface act as development centers for that grain so that it will be recognizable under the microscope.

NTA film development at Los Alamos is done according to the following procedure.⁴ Processing must be performed in a darkroom. For small scale uses, a processing hanger and tray developing is adequate. All baths are maintained at 68 °F as recommended by Kodak. Films are developed in Kodak's Rapid X-ray Developer (or developer D-19) for 5 minutes under constant agitation. The thin emulsion thickness, ~30 μm , permits agitation without significant differences in development rates at the surface and at the interior of the emulsion. The photographic developer is a weak reducing agent that is catalyzed by the silver specks. If a sufficient size development center was created during exposure, the entire grain will be reduced and thus be visually distinguishable under the

microscope; otherwise, the grain will be unaffected. The film is then placed in a running water bath and agitated for 2 minutes to discontinue the development stage. The hanger is then transferred to a Kodak fixer bath for 30 minutes. The fixer is a sodium thiosulfate solution which will wash out undeveloped grains, thus removing the excess silver and bromide ions while the developed grains remain embedded in the emulsion. The film is then treated in a final wash bath with running water for 30 minutes. The film must then be allowed to air dry in a dust-free location before reading.

The fading of the latent image in nuclear emulsions in the period between exposure and processing was observed as early as 1931 by Blau. The grain density of the track was seen to decrease as a function of time and the ambient conditions under which the film was stored. There is much in the literature suggesting atmospheric humidity and oxygen content greatly affect the rate of fading. Beiser described fading as due to the oxidation of sensitivity specks by oxygen in the presence of water described by the reaction⁵

Silver atoms that are formed by receiving an electron at the crystal surface can be re-oxidized to silver ions. Reionization of a sufficient number of silver atoms in a clump renders that grain undevelopable, resulting in a loss of latent image.

Extensive research to monitor the rate of fading while varying these controlling factors supports the theory. In these experiments, NTA films received reference neutron exposures and were then stored at a particular temperature but at different humidities or at a number of temperatures with

the relative humidity held constant. Sayed and Piesch, Becker, and Hofert and Piesch have performed such studies of fading in NTA film.^{6,7,8} In each case, the greater temperatures and higher humidities were related to increases in the rate of fading of recoil proton tracks.

Research aimed at reducing the rate of fading has been discussed in the literature as well. Sealing the film to protect it from the ambient relative humidity reduces the rate of fading. A further improvement can be achieved by drying and sealing the film in a dry nitrogen environment. This deprivation of oxygen and water is the most successful solution to prevent driving the proposed reaction in Eq. (1). Becker describes dramatic reduction in fading rates by sealing the film in polyethylene, and even greater reduction by first drying and sealing the film in nitrogen.⁷ In a similar experiment, Bartlett and Creasey observed almost no fading at 16 weeks in NTA film desiccated and sealed in dry nitrogen and stored at 58% RH.⁹ Fading studies of NTA film and the Los Alamos NTA dosimeter were performed and are elaborated upon in this report.

THE LOS ALAMOS NTA DOSIMETER

The Los Alamos NTA dosimeter is illustrated in Figure 1. Kodak NTA film is placed inside a 0.066-cm-thick cadmium pocket of the TLD Cylolac plastic holder. Whereas the cadmium was originally intended to shield the TLD card from thermal neutrons, NTA film response has been shown to be unaffected by the presence of the filter because of the ~0.7 MeV neutron energy threshold of the film. The average Cylolac thickness shielding the film is 0.25 cm. The holder is then heat sealed in a plastic pouch of 20 mils total thickness (7 mils polyester and 3 mils polyethylene each side).

Indicating silica gel crystals are also included in the pouch. The crystals change color from bright blue to white with increased water absorption and thus indicate a breached seal. A clip is placed on the dosimeter so that it may be worn by personnel similar to the TLD badge.

The NTA film is desiccated and sealed in a dry nitrogen environment. Figure 2 shows the glove box where the operations are performed. Ultra-high purity nitrogen gas (minimum purity 99.999%) with less than 3 ppm moisture is maintained in the glove box. A desiccating jar filled with Drierite indicating desiccant is used to dry the film for 64 hours inside this nitrogen environment. The dosimeter assembly and plastic pouch sealing are then completed within the glove box.

The Los Alamos NTA dosimeter is issued for a 1-month period. The dosimeter is produced less than 1 week prior to being issued. The badges are exchanged and developed within 10 days following the end of the 1-month consignment period. Hence, the maximum time between exposure and development is about 50 days.

The plastic badge holders are color coded by the issue month in order to distinguish between permanent personnel and temporary employees or visitors. In the event that the seal of the dosimeter is breached, the badge is returned for early development and the employee reissued a temporary NTA dosimeter.

CHARACTERIZATION

A. Calibration

NTA film has frequently been calibrated at Los Alamos using a PuBe-238 or a bare Cf-252 source having an average neutron energy of 4.5 MeV or 2.3 MeV, respectively. The film is irradiated in the Cylolac plastic holder using an NBS slab phantom (40 x 40 x 15 cm methyl methacrylate slab) backing. Due to the neutron energy spectrum measurements conducted at LAMPF during 1987 and the sensitive energy range of NTA film, it was concluded that the NTA dosimeter primarily measures exposure to neutrons from approximately 10-60 MeV. A response factor of 4 mrem/track/mm² was conservatively chosen based on this energy spectrum study, film response characterization experiments performed at Los Alamos, and a general consensus of the available literature.¹⁰ A background factor of 3 tracks/mm² was selected based on a 99% confidence level.

B. Fading Study

A study during 1988 indicated that serious latent image fading problems existed at Los Alamos due to high humidity conditions during heavy rainfall periods which were concurrent with the LAMPF operating period. Films placed in TLD badge holders were irradiated to 500 mrem with either PuBe-238 or bare Cf-252. Some films were developed immediately as exposure calibration references. The remaining films were periodically developed over the next 8 weeks. Within 20 days, more than 50% fading was observed for both source-type irradiated dosimeters. Approximately 90% fading was observed at 50 days, the maximum time

period between exposure and processing. The dosimeters had been stored in an office environment following irradiation; however, no relative humidity or temperature information was recorded.

In April of 1989, air sealed and unsealed NTA film badges were irradiated to 500 mrem with PuBe-238 with the NBS slab phantom backing. The PuBe-238 source was chosen over Cf-252 because the average neutron energy is closer to the actual neutron energies present at LAMPF. The exposure conditions were repeated in May for dry nitrogen desiccated and sealed dosimeters. Badges were stored in a climate controlled room, 50% RH/70°F, and in an office environment where the temperature and relative humidity were documented. Figure 3 displays the relative number of recoil proton tracks as a function of storage time for the dry nitrogen desiccated and sealed dosimeters (4 films per group) and for unsealed badges (2 films per group) stored in the office environment. Also shown is the result of the 1988 fading study for unsealed dosimeters (2 films per group) stored in a similar office environment. Table I lists the data used in Figure 3 with the standard deviation (1σ) of the points. The most significant result evident in Figure 3 is the minimal amount of fading observed at 49 days for the nitrogen dried and sealed NTA dosimeter. The presence of some fading is evidenced by the increase with storage time in the standard deviation of the mean number of tracks, as shown in Table I. In addition, the ability to read tracks in the developed film is significantly improved in the nitrogen dried and sealed dosimeter. The tracks are very clear and well defined because of little latent fading.

Ambient conditions in the office environment varied significantly

between the two time periods during which the different type of packaging methods shown in Figure 3 were stored. The unsealed dosimeter was stored in the office environment in April and May of 1989, during which time the relative humidity was on the average 20% (minimum 10%, maximum 59%). However, the desiccated and sealed dosimeter was stored in the office environment during June and July of 1989. The relative humidity was an average of 55% over this period (minimum 28%, maximum 72%). Similarly, the unsealed badges in the 1988 fading study were stored during July and August, considered to be the heaviest rainfall season in Los Alamos. These conditions of greater humidity would explain the more rapid fading rate observed for the unsealed dosimeter in the 1988 fading study relative to that measured during the 1989 fading study.

The relative number of tracks versus storage time for dosimeters stored at 50% RH/70 °F are shown in Figure 4. Table II lists the data and standard deviation for the dry nitrogen desiccated and sealed (4 films per group), air sealed (2 films per group), and unsealed dosimeters (2 films per group) used in Figure 4. The curves are results from the 1989 fading study. The dried and sealed NTA film badges again exhibited almost no fading at 49 days within the limits of the standard deviation shown in Table II. The greatly reduced fading rate for air sealed dosimeters over those not sealed is also evident.

CONCLUSIONS

The Los Alamos NTA dosimeter adequately satisfies high energy neutron dosimetry requirements at LAMPF. Sealing the film overcomes the problem of severe latent image fading. Desiccating and sealing NTA film in a dry

nitrogen environment essentially eliminates fading over the 1-month issue period of the dosimeter. The presence of indicating silica gel crystals in the packet provides acceptable assurance of the integrity of the seal. The summation of the TLD and NTA dosimeter responses agrees with spectrum unfolding code predictions. Thus, the combination of the two dosimeters effectively covers the broad neutron energy range existing at LAMPF.

TABLE I
Recoil Proton Tracks in NTA Film
Stored in Office Environment

<u>Packaging Method</u>	<u>Storage Time (Days)</u>	<u>Mean Number of Tracks/mm²</u>	<u>Standard Deviation of Mean Number of tracks/mm² (1σ)</u>
A. Desiccated and Sealed in Nitrogen (1989)	0	88.5	3.7
	4	88.8	6.2
	11	87.5	4.1
	21	89.0	3.7
	35	86.8	8.0
	49	84.0	9.1
B. Unsealed (1989)	0	82.0	4.8
	2	77.5	2.1
	7	80.0	1.4
	14	81.0	4.2
	28	71.0	5.7
	42	53.5	6.4
	56	29.5	2.1
C. Unsealed (1988)	0	97.5	0.7
	8	77.5	0.7
	21	44.0	5.7
	43	22.5	2.1
	56	5.5	2.1

TABLE II
Recoil Proton Tracks in NTA Film
Stored at 50% RH/70 °F (1989)

<u>Packaging Method</u>	<u>Storage Time (Days)</u>	<u>Mean Number of Tracks/mm²</u>	<u>Standard Deviation of Mean Number of tracks/mm² (1σ)</u>
A. Desiccated and Sealed in Nitrogen	0	88.5	3.7
	4	87.5	3.9
	11	89.8	4.0
	21	87.3	7.7
	35	87.5	9.3
	49	83.0	10.3
B. Air Sealed	0	82.0	4.8
	2	83.0	5.7
	7	83.0	4.2
	14	76.5	6.4
	28	81.5	4.9
	42	71.0	2.8
	56	46.5	3.5
C. Unsealed	0	80.0	1.4
	2	73.5	0.7
	7	70.5	3.5
	14	67.5	7.8
	28	58.0	4.2
	42	48.5	2.1
	56	18.5	2.1

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Figure 1. The LANL NTA Dosimeter.

Figure 2. Nitrogen Glove Box for NTA Film Desiccation and Dosimeter Assembly.

Figure 3. Relative number of recoil proton tracks in Kodak NTA film stored in an office environment as a function of storage time and packaging method: A, dry nitrogen desiccated and sealed (1989); B, unsealed (1989); C, unsealed (1988).

Figure 4. Relative number of recoil proton tracks in Kodak NTA film stored at 50% RH/70 °F, as a function of storage time and packaging method (1989): A, dry nitrogen desiccated and sealed; B, air sealed; C, unsealed.

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