COMPARISON BETWEEN PADC AND FNTD NEUTRON DETECTOR SYSTEMS IN BLIND TESTS

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The objective of this study was to compare a neutron dosimetry system based on polyallyl diglycol carbonate (PADC) detectors with a new system based on $\text{Al}_2\text{O}_3\text{:C,Mg}$ fluorescence nuclear track detectors (FNTD). The irradiations, performed as part of an intercomparison organized by the Physikalisch-Technische Bundesanstalt (PTB), Germany, were on a PMMA phantom with ^{252}Cf or $^{241}\text{Am-Be}$ source, usually with the phantom surface perpendicular to the radiation beam (0° angle), and with $H_p(10)$ values between 0.3 and 7 mSv. One ^{252}Cf irradiation was performed at 30° angle, and one with an additional 1 mSv gamma irradiation. The results showed an agreement between the two techniques with an average and maximum difference between PADCs and FNTDs of 1.5 and 22%, respectively, if one compares only cases of doses >1 mSv. For one of the irradiation conditions with dose of 0.9 mSv, use of the incorrect calibration factor for the FNTD (^{252}Cf instead of $^{241}\text{Am-Be}$) led to reported values ~ 2 larger than the given doses, due to low statistics in the determination of the ratio between ^6Li -doped glass and polyethylene neutron converters. Although the FNTD track analysis algorithm may need further development, the results presented here demonstrate the feasibility of the FNTD technology and indicate areas requiring improvements.

INTRODUCTION

Neutron dosimetry remains challenging because of the large variation in the interaction cross-sections used for neutron detection (mostly elastic collisions with protons and inelastic collisions with nuclei such as ^6Li and ^{10}B , resulting in secondary charged particles), the large variation in the neutron fluence to air kerma and dose equivalent (ambient or personal) conversion coefficients, and the wide neutron energy spectrum, ranging from thermal neutrons up to hundreds of MeV, depending on the neutron source and moderation⁽¹⁾.

Track-etch detectors are frequently used for passive neutron dosimetry⁽¹⁾. The technique is based on the registration of neutron-induced recoil protons and other nuclei in materials, such as polyallyl diglycol carbonate (PADC), enlarged by a chemical or electrochemical etching and quantified by optical microscopy or light scattering⁽²⁾. Advantages include the permanent track registration after etching, and the relatively flat energy response in the region between 200 keV and 10 MeV neutron energy^(3, 4). Disadvantages include the need for etching, the strong dependence of the sensitivity and background on the material properties⁽⁵⁾, which requires strict

quality control, and the fact that the detectors are not reusable.

Fluorescent Nuclear Track Detectors (FNTDs) is a new technique for neutron dosimetry, based on the registration of tracks in $\text{Al}_2\text{O}_3\text{:C,Mg}$ crystals specially grown for this purpose. Instead of chemical damage, the registration process is the radiochromic transformation of color centers in the FNTD crystal due to ionizations produced by neutron-induced charged particles⁽⁶⁾. The radiation-induced color centers are imaged by confocal microscopy⁽⁷⁾, resulting in images of the single tracks without chemical etching. A neutron dosimeter system was developed combining the FNTDs with neutron converters⁽⁸⁾, since Al_2O_3 itself is not sensitive to neutrons. Advantages include the fact that the radiochromic transformation is stable up to 600°C , but can be reversed by a thermal treatment⁽⁹⁾ or by optical erasure using a pulsed UV laser, making the detectors reusable.

The goal of this work was to compare the two technologies (PADCs and FNTDs), taking advantage of the neutron dosimetry intercomparison organized by the Physikalisch-Technische Bundesanstalt (PTB), Germany. Intercomparisons are organized

routinely and used not only for quality assurance, but also to evaluate new technologies⁽¹⁰⁾.

METHODOLOGY

PADC detectors (Table 1) used by PSI are the TASTRAK[®] (Track Analysis Systems Ltd.) in polyamide (PA) holders (Figure 1), which provide a fast neutron converter layer⁽¹¹⁾. The holder allows for ⁶LiF chips to be placed on two opposite sides of the PADC as thermal neutron converters (currently not used in the dose calculation). After irradiation the detectors were etched using a 6.25 M NaOH solution for 2 h 50 min at 85°C.

The FNTDs (Table 1) consists of Al₂O₃:C,Mg single crystals placed on a holder (Figure 1) in contact with two neutron converters, polyethylene (PE) for fast neutrons and ⁶Li-loaded glass for thermal neutrons, as well as a Teflon[®] (PTFE) for gamma signal subtraction⁽⁸⁾. The holders also contain three positions for optically stimulated luminescence detectors (not used). Before assembly, the FNTDs were bleached with a pulsed UV laser and read to ensure low residual background (<1 track/mm²) and good surface quality.

Irradiations were performed in the PTB irradiation room (7 m × 7 m × 6.5 m) at 3.25 m above ground with the detectors on the surface of a 30 cm × 30 cm × 15 cm PMMA phantom, using either a ²⁵²Cf source or a ²⁴¹Am–Be source (see nominal conditions in Table 3). One ²⁵²Cf irradiation was performed with a 30° angle and one set of detectors was given an additional gamma dose from a ¹³⁷Cs source.

The PADCs and FNTDs were calibrated at PSI's accredited laboratory with ²⁴¹Am–Be $H_p(10) = 3$ mSv in reference conditions, on the surface of a 30 cm × 30 cm × 15 cm ISO water phantom.⁽¹²⁾ The calibration was also verified by irradiation with ²⁴¹Am–Be at Landauer Inc. (Glenwood calibration laboratory) with 5 mSv, with 3 mSv bare ²⁵²Cf irradiation at PSI, and with 1 mSv and 3 mSv bare ²⁵²Cf irradiation at SCK-CEN Belgium.

PADC detectors were analyzed using the TASLIMAGE Neutron Dosimetry System (Track Analysis Systems

Ltd.) in the same conditions used for routine dosimetry at PSI. The system performs automatic track analysis and calculates the track density after rejection of artefacts based on a proprietary algorithm. The dose is calculated by simple comparison with the track density of calibration detectors. For the used PADC batch, the calibration factor was 400 tracks cm⁻² mSv⁻¹. Each detector analysis takes ~30 s. The ratio between the track counts under the ⁶LiF chip and the PA converters (Li/PA ratio) were determined by an independent scan.

The FNTDs were analyzed at Landauer, Europe using a confocal laser scanning microscope system developed by the company and proprietary analysis algorithm⁽⁸⁾. Result for the calibration detectors are shown in Table 2. The ratio between the Li-loaded glass and PE is referred to as Li/PE ratio. The FNTD response under different converters is dependent on the neutron energy⁽¹³⁾. Based on the data of Table 2 and previous results, the calibration factor for the ²⁴¹Am–Be spectrum was adopted if the Li/PE ratio was < 0.4, and for the ²⁵²Cf spectrum if the Li/PE ratio was ≥ 0.4. If the Li/PE ratio < 0.5

Figure 1. PADC and FNTD detectors used in this study.

Table 1. Characteristics of the PADC detectors and FNTDs used in this study.

	PADC	FNTD
Dimensions	20.0 mm × 25.0 mm × 1.5 mm	4.0 mm × 8.0 mm × 0.5 mm
Manufacturer	Track Analysis Systems Ltd.	Landauer Inc.
Converters	2 mm polyamide 3 mm × 3 mm × 0.9 mm ⁶ LiF chips	⁶ Li-loaded glass: (830 ± 25) µm Polyethylene (PE): 2 mm PTFE
Preparation	—	Bleached with pulsed UV laser bleaching
Post-irradiation etching	6.25 M NaOH solution for 2 h 50 min at 85°C	—
Scanned area	1.0 cm ²	2.76 mm ² per converter

and the CV of the ratio $> 30\%$ we considered the energy determination as 'uncertain' (see irradiation #16, Table 3) and used the ^{252}Cf calibration factor.

RESULTS AND DISCUSSION

Table 3 summarizes the results for the various irradiation conditions. The FNTD track densities were $\times 2.1$ higher than the PADC track densities for the ^{252}Cf neutron spectrum, and $\times 4.8$ higher for the ^{241}Am -Be neutron spectrum. These results indicate the strong energy dependence of FNTD in comparison with PADC, in agreement with the calibration data (Table 2) and the energy response of these detectors^(3, 13). The data demonstrates an agreement within $\pm 22\%$ between the PADCs and FNTDs, except for doses < 1 mSv (irradiation #15 and 16, see more below).

The ratios between reported and delivered doses were similar for PADCs and FNTDs, with deviations within $\pm 30\%$, except irradiation #15 because of the low dose, and irradiation #16 in the case of the reported FNTD result; using the correct calibration factor, the result agree with the PADC results. Furthermore, the additional 1 mSv gamma dose in irradiation #11 did not cause a problem for either PADCs or FNTDs.

For unirradiated PADC detectors, no dose was observed, indicating the good performance of the TASLIMAGE system background discrimination algorithm (data not shown). For the FNTDs, the mean dose of the unirradiated detectors was 0.08 mSv, with a standard deviation of 0.09 mSv. For a single FNTD a dose of 0.28 mSv was reported. It is not clear at this point if this is due to the influence of cosmic rays during transport or detector fluorescence background.

Table 2. FNTD calibration for irradiation in various facilities.

Source	Dose (mSv)	Sensitivity of PE converter area (tracks cm^{-2} mSv $^{-1}$)	Li/PE	sd Li/PE
Am-Be (PSI)	3	1740	0.24	0.14
Am-Be (Landauer)	5	1960	0.27	0.03
^{252}Cf (PSI)	3	824	0.85	0.11
^{252}Cf (SCK-CEN)	3	835	0.52	0.25

Table 3. PADC and FNTD results for the different irradiation conditions (track densities behind fast neutron converter).

Irradiation # source, angle, distance	H_i (mSv)	PADC track density (tracks cm^{-2})	FNTD track density (tracks cm^{-2})	FNTD energy	PADC H_m (mSv)	FNTD H_m (mSv)	PADC H_m/H_i	FNTD H_m/H_i
11 $^{252}\text{Cf} + ^{137}\text{Cs}$, 0°, 58 cm	1.00 (5) (+1 mSv γ)	481	850	^{252}Cf	1.2	1.02	1.20	1.02
		421	1060	^{252}Cf	1.05	1.28	1.05	1.28
		474	950	^{252}Cf	1.18	1.15	1.18	1.15
		369	820	^{252}Cf	0.92	0.99	0.92	0.99
12 ^{252}Cf , 0°, 58 cm	5.00 (24)	2147	4260	^{252}Cf	5.36	5.13	1.07	1.03
		2111	4750	^{252}Cf	5.27	5.72	1.05	1.14
		2079	4460	^{252}Cf	5.18	5.37	1.04	1.07
		2097	4490	^{252}Cf	5.24	5.41	1.05	1.08
13 ^{252}Cf , 0°, 100 cm	7.00 (22)	3092	6380	^{252}Cf	7.72	7.69	1.10	1.10
		3002	6300	^{252}Cf	7.50	7.57	1.07	1.08
		3285	6610	^{252}Cf	8.20	7.97	1.17	1.14
		2993	5800	^{252}Cf	7.47	6.94	1.07	0.99
14 ^{252}Cf , 30°, 100 cm	3.50 (11)	1063	2300	^{252}Cf	2.65	2.81	0.76	0.80
		1108	2350	^{252}Cf	2.77	2.84	0.79	0.81
15 ^{252}Cf , 0°, 100 cm	0.30 (1)	98	330	^{252}Cf	0.24	0.40	0.80	1.33
		121	290	^{252}Cf	0.30	0.35	1.00	1.17
		132	480	^{252}Cf	0.33	0.58	1.10	1.94
		63	220	^{252}Cf	0.16	0.27	0.53	0.90
16 ^{241}Am -Be, 0°, 100 cm	0.90 (5)	344	1560	Uncertain	0.86	1.88*	0.96	2.09 ^a
		316	1610	Uncertain	0.79	1.94*	0.88	2.15 ^a

^aDose calculated using calibration for ^{252}Cf . The calculation was revised using calibration for ^{241}Am -Be source and the results were 0.90 and 0.92 mSv.

Figure 2 shows The Li/PA and Li/PE ratios. Besides the difference in magnitude and the large uncertainties, similar trends were observed for PADCs and FNTDs. The PADC Li/PA ratio was in general >8 , and varying with irradiation conditions. Normally the PADC Li/PA ratio is ~ 1.0 for irradiation with fast neutrons without phantom, but for irradiations on phantom the track density behind ${}^6\text{LiF}$ chip is 10 times higher than behind PE converter because of the neutrons thermalized by the phantom.

For the FNTDs, the Li/PE ratio varied between 0.14 and 1.00, but in most cases were >0.4 . Because of that, most of the dose calculations assumed the ${}^{252}\text{Cf}$ calibration. The detectors in irradiation #16 did not provide a sufficient precise estimate for the Li/PE ratio due to low statistics and, therefore, the energy was classified as ‘uncertain’ and the doses were reported using the ${}^{252}\text{Cf}$ calibration, resulting in a discrepancy of a factor of 2. These results were later revised with the ${}^{241}\text{Am-Be}$ calibration, resulting in agreement with PADC results. Larger FNTD areas could be scanned to improve the counting statistics at low doses.

CONCLUSIONS

The results presented here provide the first independent comparison between PADC and FNTDs. The results showed an agreement between the two techniques, with an average and maximum differences between PADCs and FNTDs of 1.5 and 22%, respectively, if one compares only cases in which the doses were > 1 mSv. For one of the irradiation with dose < 1 mSv, use of the incorrect calibration factor (${}^{252}\text{Cf}$ instead of ${}^{241}\text{Am-Be}$) led to reported values $\sim \times 2$ larger than the given doses for the FNTDs. This was

Figure 2. Ratio between the track density behind ${}^6\text{Li}$ converter and PA or PE converter for PADCs and FNTDs, as a function of irradiation condition.

caused by the low statistics in the determination of the neutron energy spectrum based on the ratio between ${}^6\text{Li}$ and polyethylene neutron converters, which can be improved by scanning larger FNTD areas. The background discrimination algorithm from the TASLIMAGE system showed good performance, whereas for one unirradiated FNTDs a dose of 0.28 mSv was reported. It is not clear if this was due to the influence of cosmic rays during transport or to the background in the detectors. This study represents only a small set of data—PADCs can also lead to false positives if material problems are not properly handled by the track filtering algorithm.

Although the FNTD track analysis algorithm may need further development, these results provide evidence of the FNTD technology feasibility for neutron dosimetry and indicate areas where improvements are required. At the moment the main disadvantages of the FNTDs in comparison with PADCs are the readout time and the energy response. FNTD measurements tend to sample smaller surface area compared to PADCs, which may lead to larger uncertainties. Enlarging the FNTD scan area can be seen as a major area of improvement. On the one hand, the FNTD readout time is much longer than the PADC readout time, but on the other hand no etching is required. The FNTD energy dependence also requires precise determination of the Li/PE ratio, and therefore a larger scanned area for better statistics. If these technical issues are addressed, the main advantage of the FNTDs will be the possibility of re-using the detectors in passive neutron dosimetry.

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