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Preliminary Characterization of the Passive Neutron Dose Equivalent Monitor with TLDs

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Abstract

The passive neutron dose equivalent monitor with TLDs is composed of a cubic polyethylene moderator and TLDs at the center of moderator. This monitor was originally designed for measurements of neutron doses over long-term period of time around the nuclear facilities.

In this study, the energy response of this monitor was calculated by Monte Carlo methods and experimentally obtained under ^{241}Am-Be, ^{252}Cf and moderated ^{252}Cf neutron irradiation. Additionally, the responses of two types of conventional neutron dose equivalent meters (rem counters) were also investigated as comparison.

The authors concluded that this passive neutron monitor with TLDs had a good energy response similar to conventional rem counters and could evaluate neutron doses within 10 % of accuracy to the moderated fission spectra.

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TLD 内蔵中性子線量当量計の特性評価

（研究報告）

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要旨

TLD を内蔵した積算型中性子線量当量計の中性子エネルギー特性を調査した。

この線量当量計は、立方体状のポリエチレン減速材とその中心位置の TLD から構成され、長期間にわたる積算中性子線量の測定を目的に設計されている。

本研究では、この TLD 内蔵中性子線量当量計について、その中性子エネルギー応答特性をモンテカルロ計算及び ^{241}Am-Be、^{252}Cf 及び ^{252}Cf の減速中性子校正場における照射実験から評価した。一般的に使用されている既存の中性子線量当量計（レムカウンタ）と比較して、本 TLD 内蔵中性子線量当量計は同等のエネルギー応答特性を持っており、上記の中性子場に対して10%程度の不確かさで線量を評価できる性能を有していることが明らかとなった。

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1. Introductions

By thermalizing the neutrons in a hydrogenous moderator, Anderson and Braun developed a neutron detector with a good response to all energies up to 10 MeV [1]. Their instrument was composed of a cylindrical polyethylene moderator with inner neutron absorber shell and a thermal neutron detector ($^{10}BF_3$ proportional counter) at the center of moderator. These types of neutron survey instruments are generally called "rem counter". A various type of rem counters is commercially available and have been widely used for measurements of neutron doses at the workplaces of nuclear facilities.

In most existing rem counters, active proportional counters filled with $^{10}BF_3$ or ^{3}He gases are used as a thermal neutron detector. However, these type of rem counters are not suitable to get the dose equivalent over a longer period of time or to determine the average dose equivalent rate in a pulsed neutron field where proportional counters suffer from dead time losses. Therefore, in such neutron fields, the passive type of rem counters is required.

Recently, a new passive rem counter using TLDs has been developed by Uwamino et al.[2] and commercially available from Panasonic Ltd. This instrument consists of a simple cubic polyethylene moderator and TLDs and originally designed for the purpose of measurements of neutron doses over long-term period of time around the nuclear facilities.

The authors are planning to implement this instrument in the radiological controlled area for measurements of the burst neutrons in case of unexpected criticality accidents because this instrument can essentially prevent from suffering dead time losses. In order to evaluate the performance of this neutron dose equivalent monitor with TLDs, some experiments to obtain its neutron energy response was carried out. At the same time, some conventional active types of neutron dose equivalent meter (rem counters) were also used as comparison.

2. Material and Methods

2.1 Neutron Dose Equivalent Monitors

Neutron dose equivalent monitor with TLDs

The neutron dose equivalent monitor with TLDs (TLD rem counter) used under this study consists of two parts, a moderator and TLDs.

The moderator is Panasonic UD-893 "DICE" moderator, shown in *Figure 1.* It is a cubic polyethylene moderator with the dimension of 22cm with inner Cd absorbers to improve neutron energy dependence. At the center of moderator, the chamber made of lead is placed and two same TLDs will be contained horizontally with one vertical to the other like a cross.

The TLDs used with this instrument is the Panasonic UD-813PQ4 shown in *Figure 2.* Four thermoluminesecent phosphors are housed in a plastic holder listed below.

Element Number	Phosphor Material	Filter in TLD
1	^{6}Li2^{10}B4O7(Cu)	160mg/cm^{2}, plastic
2	^{6}Li2^{10}B4O7(Cu)	
3	^{7}Li2^{11}B4O7(Cu)	
4	^{7}Li2^{11}B4O7(Cu)	

The UD-813PQ4 consists of one holder and one plate. The holder is for slotting in the plate, while the plate is the basement of TL elements. Four TL elements are enriched lithium borate phosphors, two ^{6}Li2^{10}B4O7(Cu) and two ^{7}Li2^{11}B4O7(Cu). These four TL elements are inlayed in the plastic plate one by one.

The moderated neutrons in a polyethylene moderator are captured by ^{6}Li(n,alpha) or ^{10}B(n,alpha) reactions in ^{6}Li2^{10}B4O7(Cu) phosphors. Since ^{7}Li2^{11}B4O7(Cu) phosphors are not sensitive to neutron, the net reading is obtained by subtraction of both readings as follows.

N = (average readings of ^{6}Li2^{10}B4O7(Cu)) – (average readings of ^{7}Li2^{11}B4O7(Cu))

Conventional neutron dose equivalent meters

Two kinds of conventional rem counters, cylindrical type (Studsvik/Alnor 2202D) and spherical type (Fuji electric Co., Ltd., NSN1) were chosen for comparison. Both rem counters have been used for radiation control in MOX fuel fabrication facilities of Tokai Works.

2.2 Neutron Sources and Neutron Reference Calibration Field

Neutron irradiation experiments were carried out at the Calibration Facility for Radiation Monitoring Instruments, Tokai Works. This calibration facility is in charge of inspecting, calibrating and repairing radiation monitoring instruments used in Tokai Works and equipped with many standard radioactive sources and irradiation apparatus.

In our experiment, we used two kinds of neutron standard sources, ^{241}Am-Be and ^{252}Cf. The neutron emission rates from ^{241}Am-Be and ^{252}Cf were evaluated with good accuracies by the Electro-Technical Laboratory, Tsukuba, and the National Physics Laboratory, UK, respectively. The uncertainty of emission rate is 4% for ^{241}Am-Be and 0.6% for ^{252}Cf. The neutron fields produced by using these sources are internationally standardized fields and widely used for calibration of neutron survey instruments and personal monitors.

In addition, we also used moderated neutron fields for the purpose of the performance evaluation of the rem counters in the workplaces of MOX fuel fabrication facilities. The moderated neutron fields were established by surrounding a ^{252}Cf standard source with lead-contained PMMA moderators shown in *Figure 3*. There is a set of four hollow cylindrical moderators and its thickness is changeable from 13.5mm to 77.5mm by combining each moderator. A lead-contained PMMA is a common shielding material of the glove-boxes and therefore this field is a good simulator for real working condition in MOX fuel fabrication facilities.

The neutron irradiation apparatus is installed at the irradiation room B of the calibration facility. This room has a floor size of 6m x 6m with a 2 m depth underground pit. The wall and roof are made of a low-density concrete for reducing the room reflected neutrons. The neutron source is pushed up by an air pressure along with a guide-transfer tube from an underground storage container and placed at irradiating position above 1.2 m from the grating-hatch. *Figure 4* shows the irradiating system, where a transfer tube is surrounded with moderators.

The neutron energy spectra of these neutron fields were evaluated from calculation using Monte Carlo neutron transport code, MNCP4B [3], and also verified from experiments. In the calculations, the contribution of room-scattered neutrons was fully considered. *Figure 5* shows the calculated neutron spectra at the reference calibration point (1 m away from the source) [4]. The calculated neutron spectra are in good agreement with the spectra measured by BSS (Bonner Sphere Spectrometer).

The dosimetric parameters at the reference calibration point are summarized in *Table 1*. All parameters include (direct and penetrating) neutron components and room-scattered components. The ambient dose equivalent, *H*(10)*, delivered in this irradiation experiments were obtained from the following equation.

$$H*(10) = \overline{h*(10,r)} \times B \times T$$

Where, $h*(10,r)$ is a neutron emission rate to ambient dose equivalent conversion factor at a distance of *r*, listed in *Table 3*. ***B*** and ***T*** are a neutron emission rate and irradiation time, respectively.

2.3 Experimental Configuration

Figure 6 is the layout for experiments. The rem counteres were placed on a calibration pushcart. The distance between the sensitive center of rem counters and source was fixed to 1 m by using the laser-positioning system.

The Irradiation distance and time are summarized in *Table 2*. The neutron emission rate at the date of experiments was 1.87E+7 s^{-1} for ^{252}Cf and 2.33E+6 s^{-1} for ^{241}Am-Be.

3. Calculation of Response Function of TLD

The response function of TLD rem counter cannot be determined only from the experiments because mono-enegetic neutrons which can be available for calibration are limited to thermal and above about 100 keV. We therefore calculated its response function and the calculational accuracy was verified by comparing with experiments.

A Monte Carlo method is applied for this calculation because the three-dimensional geometric configuration of TLD rem counter is so complicated. Calculation conditions are listed below.

Transport Code	: MCNP4B
Data Library	: ENDF/B-6
Geometric configurations	: Geometries of polyethylene moderator, inner Cd absorbers and TLD containing lead-chamber are fully simulated
Source	: Mono-energetic neutron and thermal maxellian beam, perpendicularly and/or randomly striking into the face of moderator

The neutron flux averaged over the TLD position, at the center of moderator, was estimated by the spherical volume detector (*tally F4*) with a diameter of 10 mm and the intensity of thermoluminesence was calculated by a convolution between the calculated neutron spectrum and the theoretical neutron response function of $^{6}Li2^{10}B4O7(Cu)$ in a free-air.

The response function of TLD rem counter to neutron with an initial energy of ***En*** was obtained from the following calculation.

$$R(En) = \frac{\int \phi_{LBO}(En \rightarrow E) \cdot R_{LBO}(E)dE}{\Phi(En)}$$

where,

$R(En)$	: Response function of TLD rem counter to neutrons, with initial energy of ***En***, striking into the face of moderator
$\phi_{LBO}(En \rightarrow E)$	: Neutron fluence with energy of ***E***, at the TLD position
$R_{LBO}(E)$	: Response function of $^{6}Li2^{10}B4O7(Cu)$ in a free-air, calculated by Iwai (5)
$\Phi(En)$	: Neutron fluence, striking into the face of moderator

Figure 7 shows the comparison of experimental[2] and calculated response functions of TLD rem counter. Those results are expressed as mR/(n cm^{-2}), where mR is a read unit of thermoluminescent intensity by readers. The fluence to ambient dose equivalent conversion coefficients, quoted from ICRP Publication 74 [6], are also superimposed in this figure in arbitrary unit. The calculated results are also listed in *Table 3*.

The calculated response function is in a good agreement with experimental one in the high energy region. The energy dependence of response curve is also very close to that of the fluence to ambient dose equivalent conversion coefficients in the energy region from 0.1 MeV to several MeV. For the intermediate energy region, this TLD rem counter may give an overestimation of doses by up to a factor of 5, however, these situations are quite specific. Since the typical neutron spectra encountered in nuclear facilities have fission spectra around a few MeV and 1/E slowing-down spectra below a few hundred keV due to hydrogen elastic scattering, those overestimation can be compensated.

4. Experimental Results

The TLD rem counter was irradiated to the six kinds of neutron fields, ^{241}Am-Be, ^{252}Cf and moderated ^{252}Cf. TLD rem counter contains two TLDs and totally twelve TLDs were used for irradiation. After irradiation, an automatic TLD reader, Panasonic UD-7100P, read out the received doses from TLDs. TLD readers is calibrated for indicating photon dose in unit of roentgen equivalent, mR eq., therefore, we must determine the calibration factor from the readings to the real neutron dose.

The response to various neutrons, ***R***, were obtained as follows.

$$R=N/H$$

Where, ***N*** is a net reading of TLDs to neutrons and ***H*** is delivered dose.

The inverse of ***R*** corresponds to the calibration factor, ***K***.

$$K=R^{-1}$$

The experimental results were summarized in *Table 3*. The calibration factors obtained in the six irradiations has almost constant values, 0.297 to 0.344.

Here, we determine the value of 0.337 to ^{252}Cf as the default calibration factor because ^{252}Cf source is widely used as a calibration source. By multiplication of the default calibration factor to all experimental results, we can plot out the value of measured dose/delivered dose as a function of average neutron energy in *Figure 8*. This figure shows that the energy dependence of TLD rem counter is very small and it can measure neutron doses in the moderated fission neutron fields with a good accuracy.

Additionally, the response of two conventional rem counters are also summarized in *Table 4* and its responses are plotted in *Figure 9* as a function of average neutron energy. The energy response of TLD rem counter is very similar to those of two conventional rem counters.

5. Conclusion

The characteristics of this monitor based on the present study can be summarized as follows.

(1) The neutron energy response of TLD rem counter to mono-energetic neutrons was calculated by a Monte Carlo method. The calculated response curve is very close to that of the fluence to ambient dose equivalent conversion coefficients curve, presented by ICRP Publication 74, in the energy region from 0.1 MeV to several MeV.

(2) The response of TLD rem counter was investigated under irradiation experiments at ^{241}Am-Be, ^{252}Cf and moderated ^{252}Cf neutron calibration fields. It is verified that TLD rem counter is a good and suitable neutron dose equivalent monitor for MOX fuel fabrication facilities.

Additionally, compared to the conventional active neutron dose equivalent meters, TLD rem counter has the following advantages.

- Low costs
- No electric power supply
- Suitable for long-term and/or low dose measurements
- Suitable for pulsed neutron field

Therefore, the authors are sure that TLD rem counter will be served for routine periodical neutron monitoring around the vicinity of the radiological controlled area and for retrospective dosimetry in case of unexpected criticality accidents.

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[2] Uwamino,Y. et al.; *Development of a Neutron Dose Equivalent Monitor with TLB(2)*, Proceedings of the Fall Meeting of Japan Atomic Energy Society (1997)
[3] Briesmeister,J.F. ; LA-12625-M, (1997)
[4] Tsujimura,N. et al. ; *Development of Neutron Reference Calibration Field using* ^{252}*Cf and Hollow Cylindrical PMMA Moderators*, Proceedings of the Fall Meeting of Japan Atomic Energy Society (2000)

[5] Iwai,S.,; private communication
[6] ICRP ; ICRP Publication 74, (1997)

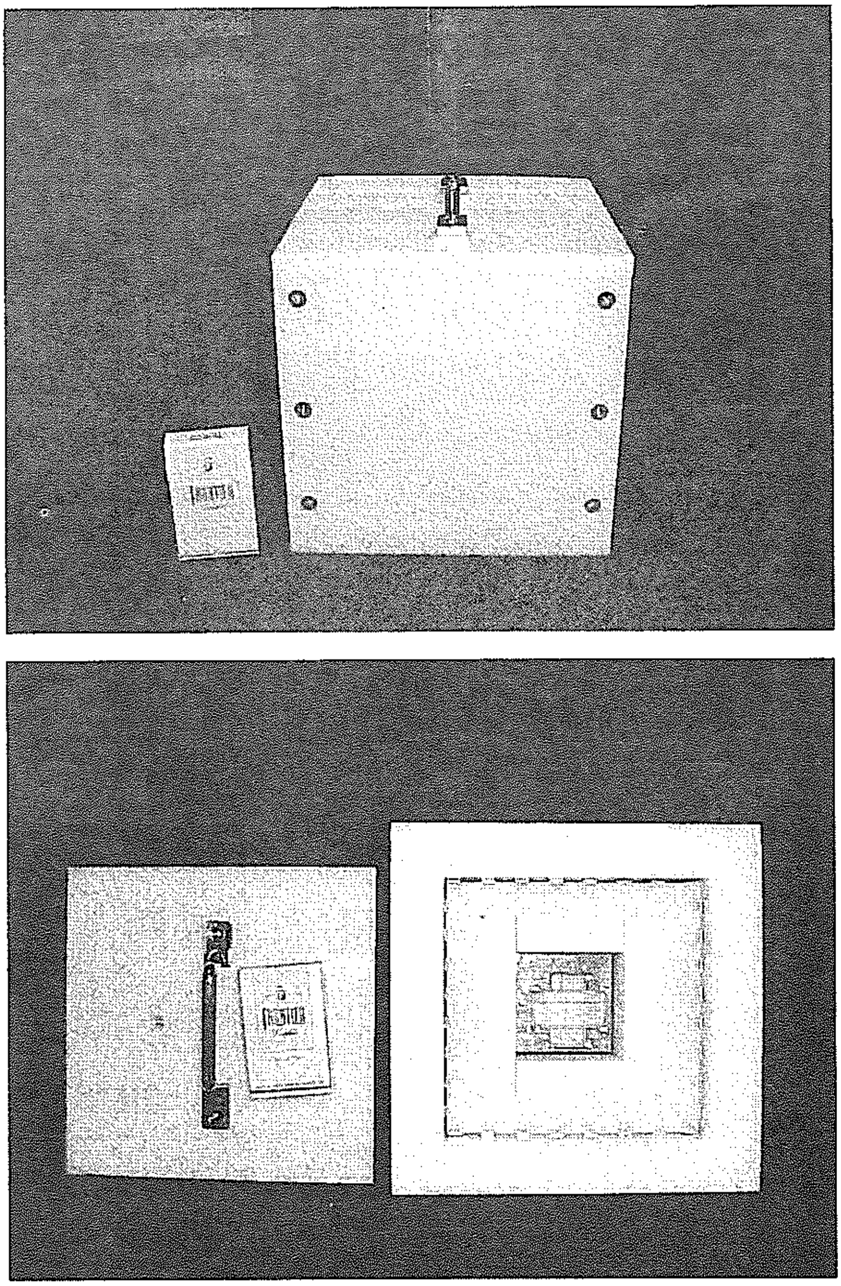

Figure 1 External view of Panasonic UD-813P neutron moderator

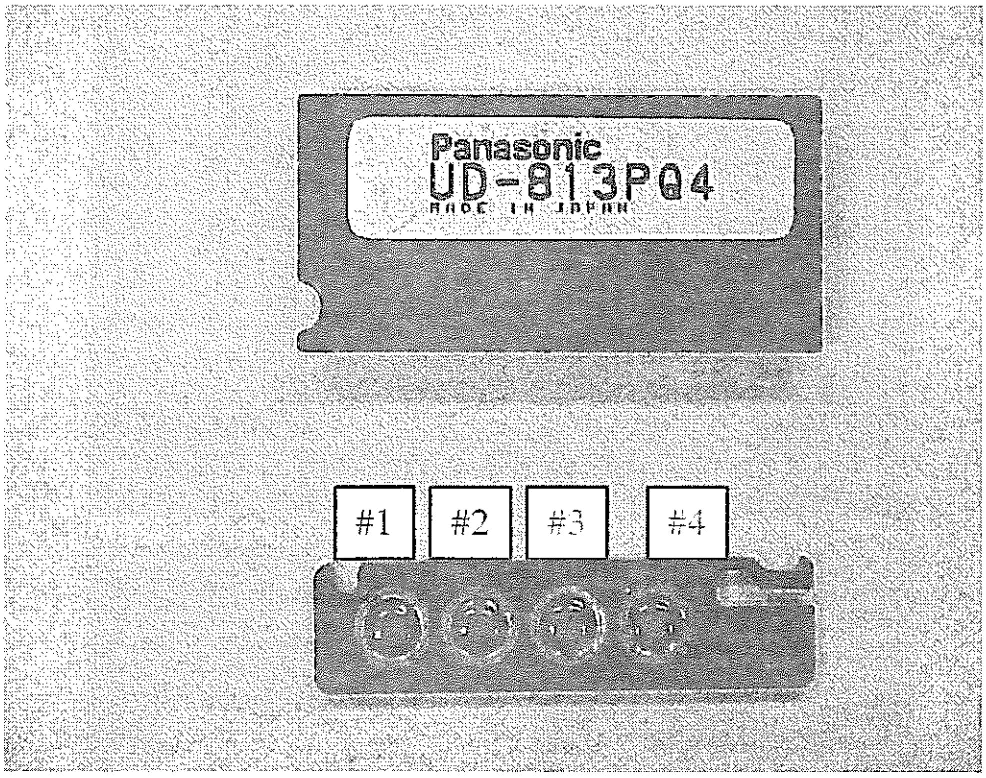

Figure 2 External view of Panasonic UD-813PQ4. Element 1,2 are $^{6}Li2^{10}B4O7(Cu)$ and element 3,4 are $^{7}Li2^{11}B4O7(Cu)$.

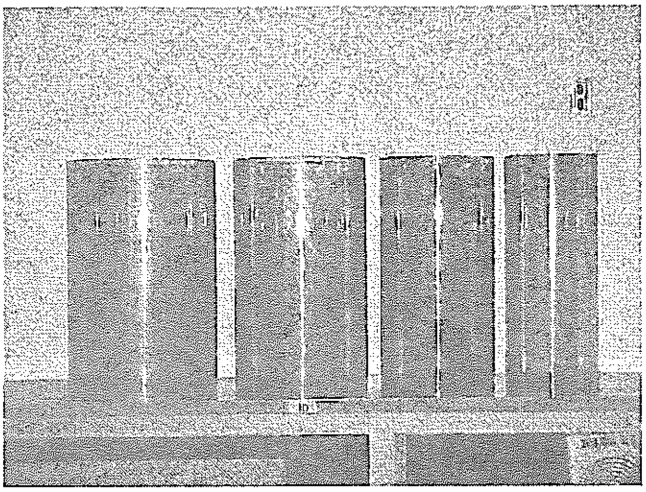

Figure 3 Hollow cylindrical PMMA moderators
($1.6 g/cm^3$, 50cm height)

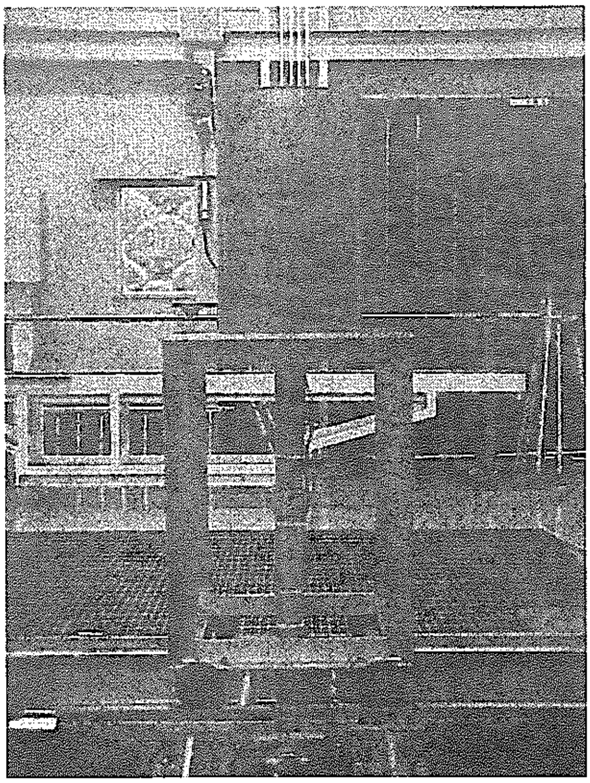

Figure 4 Neutron Irradiator, where PMMA moderators surround a transfer tube

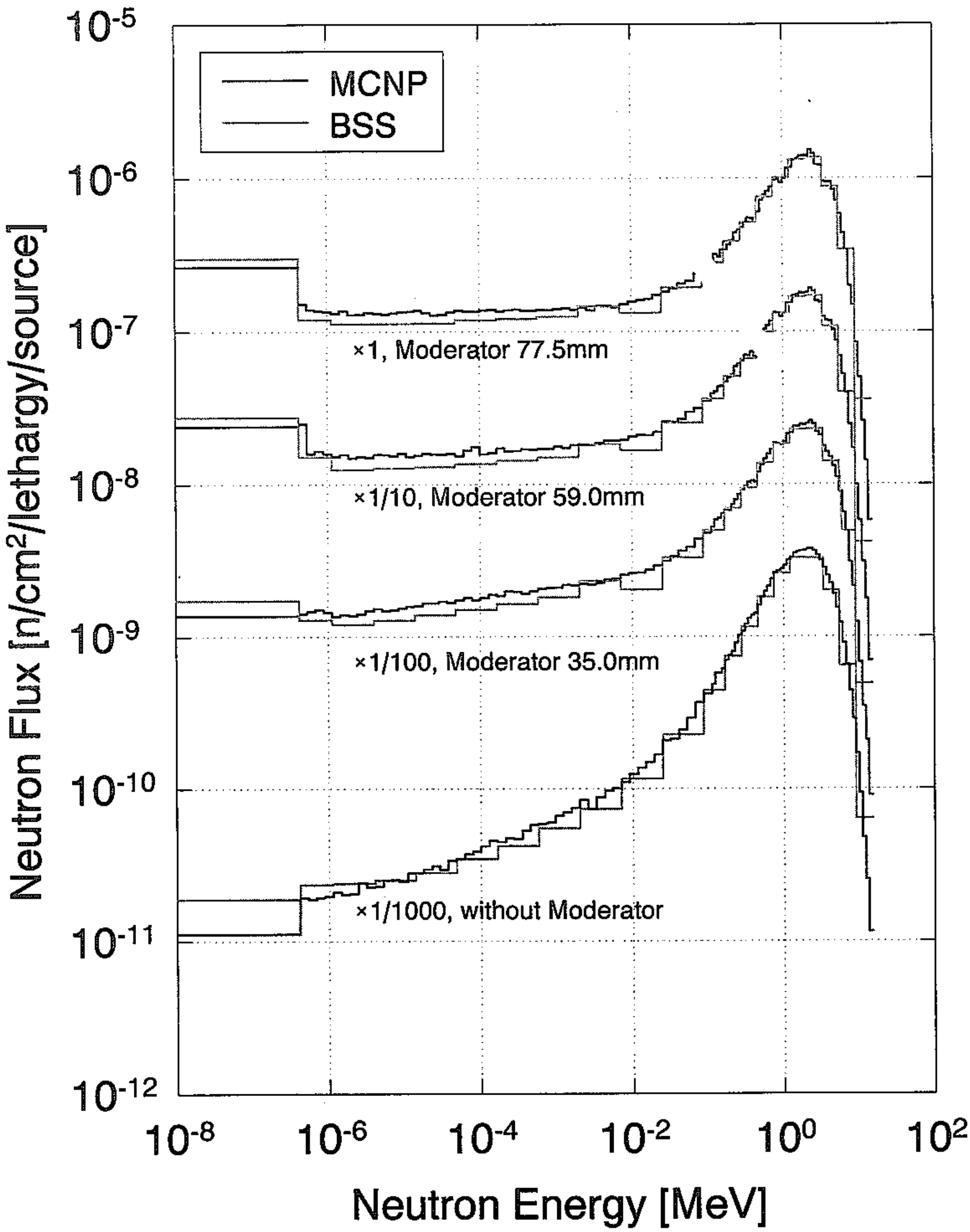

Figure 5 Neutron energy spectrum at the reference calbration point

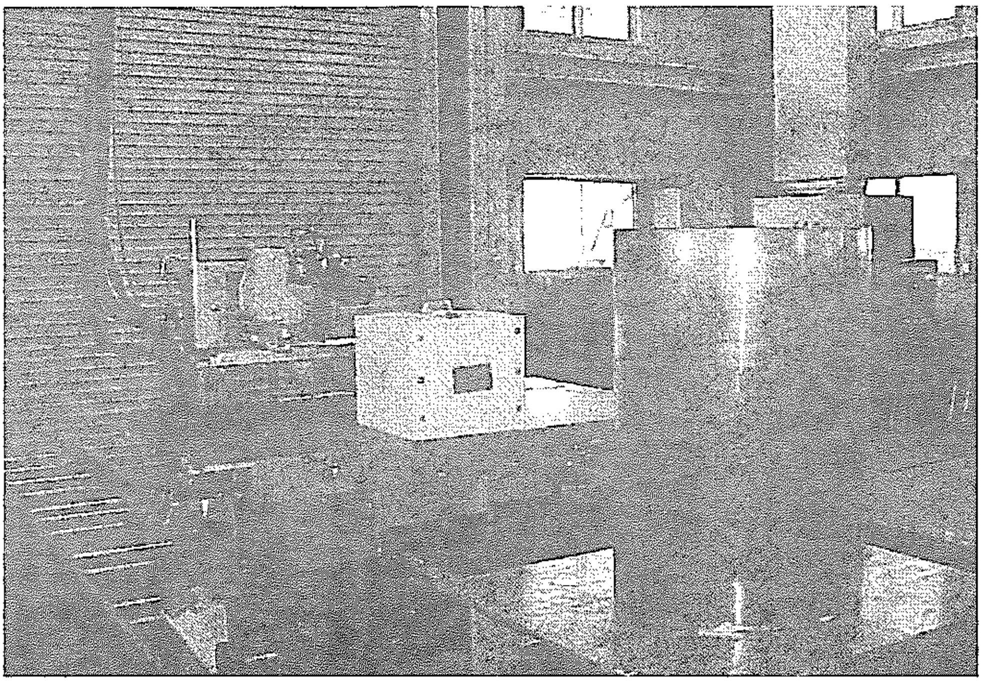

Figure 6 Experimental set-up

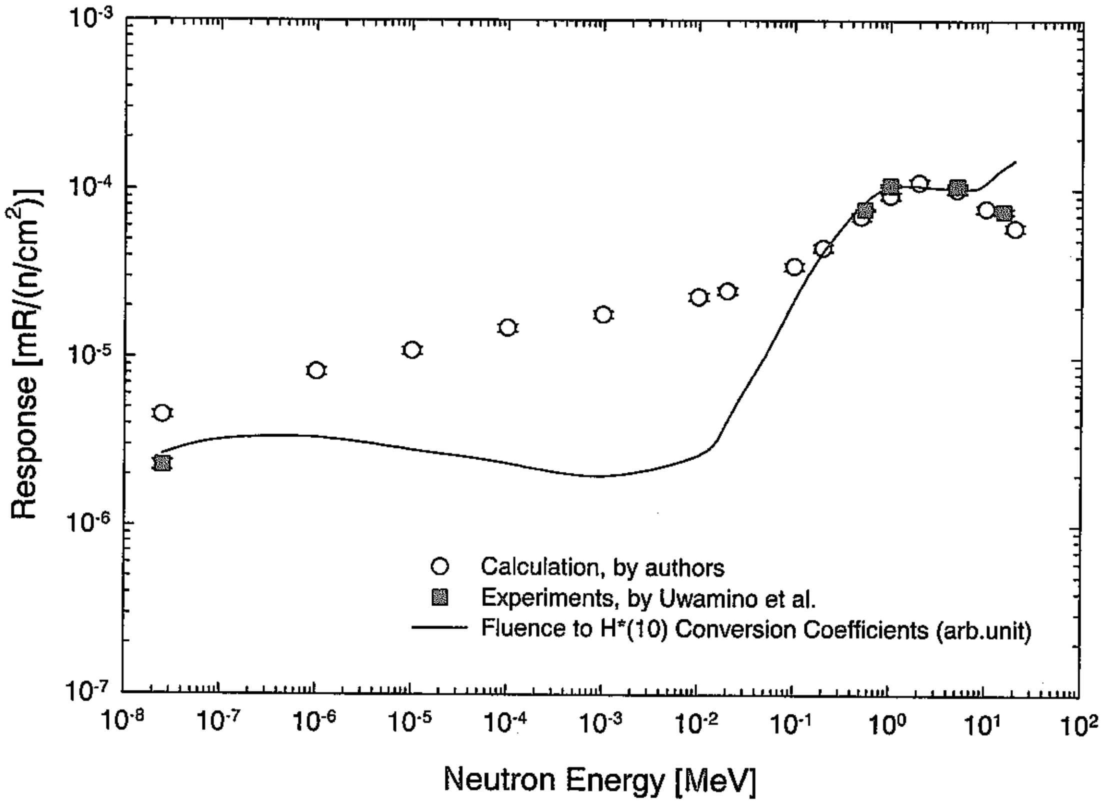

Figure 7 Response function per unit neutron fluence of TLD rem counter

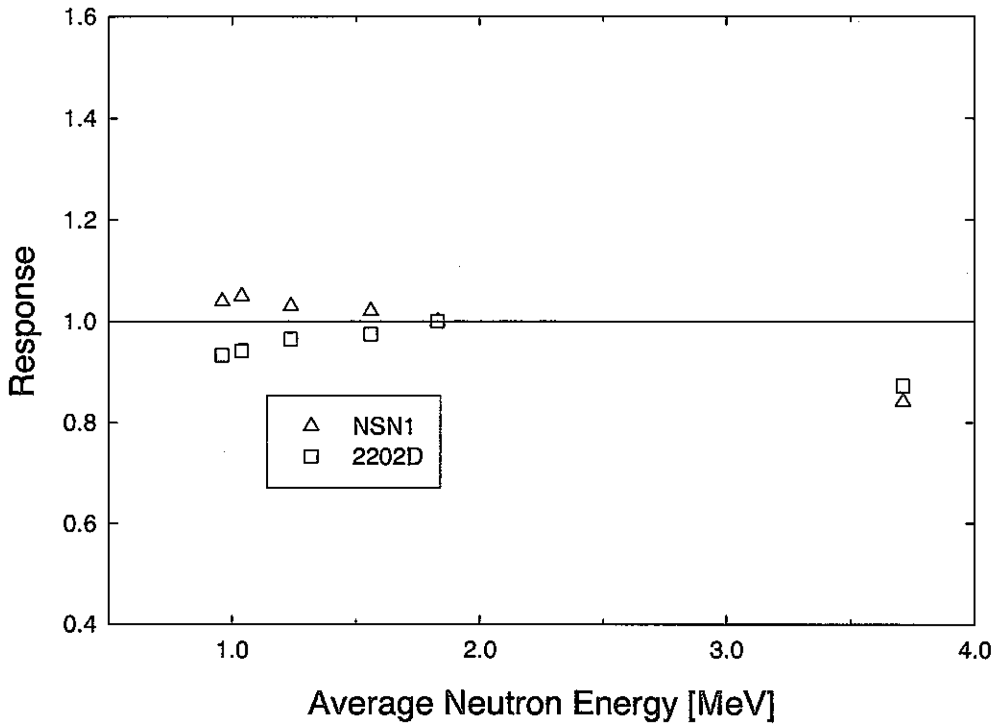

Figure 8 Response of TLD rem counter as a function of avareged neutron energy. The values are set to 1 at ^{252}Cf.

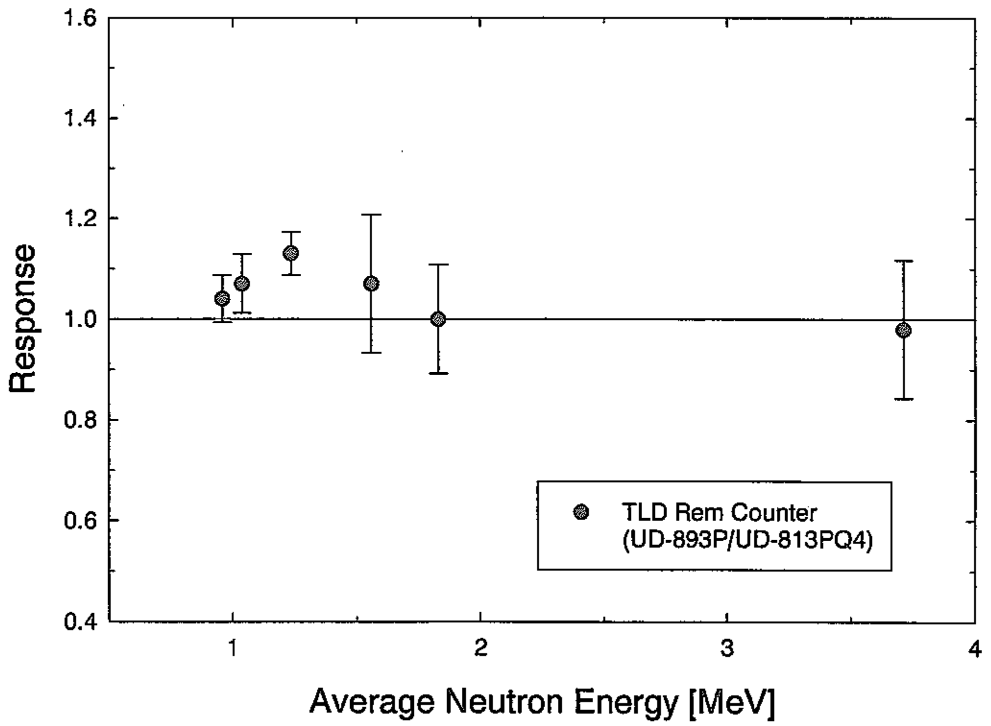

Figure 9 Response of conventinal rem counter as a function of avareged neutron energy. The values are set to 1 at ^{252}Cf.

Table 1 Dosimetric Parameters at reference calibration point (100cm from source)

		^{252}Cf					$^{241}Am\text{-}Be$
		Without Moderators	Moderators 13.5mm	Moderators 35.0mm	Moderators 59.0mm	Moderators 77.5mm	Without Moderators
Fluence[n/cm^2/source]		9.86E-06	1.02E-05	9.97E-06	8.79E-06	7.50E-06	1.85E-05
$h^*(10)$ [pSv/source]	ICRP Publ.51	2.85E-03	2.56E-03	2.00E-03	1.47E-03	1.14E-03	3.14E-03
	ICRP Publ.74	3.32E-03	2.99E-03	2.34E-03	1.71E-03	1.33E-03	3.42E-03
Average Energy [MeV]		1.83 [1)]	1.56	1.24	1.04	0.97	3.71
		2.22 [2)]	2.16	2.16	2.20	2.24	4.17
		1.91 [3)]	1.85	1.85	1.88	1.93	3.84

1) Neutron energy averaged over the spectrum at the reference point

2) Neutron energy averaged over the ambient dose equivalent (ICRP Publ.51) spectrum at the reference point

3) Neutron energy averaged over the ambient dose equivalent (ICRP Publ.74) spectrum at the reference point

Table 2 Irradiation parameters

	TLD rem counter		Fuji rem counter		Studsvik rem counter	
	Irradiation distance* (m)	Irradiation time (Minutes)	Irradiation distance (m)	Counting time (Minutes)	Irradiation distance (m)	Counting time (Minutes)
^{252}Cf	1.0	100	1.0	1.0	1.0	1.0
^{252}Cf +PMMA 13.5mm	1.0	100	1.0	1.0	1.0	1.0
^{252}Cf +PMMA 35.0mm	1.0	100	1.0	1.0	1.0	1.0
^{252}Cf +PMMA 59.0mm	1.0	104	1.0	1.0	1.0	1.0
^{252}Cf +PMMA 77.5mm	1.0	100	1.0	1.0	1.0	1.0
^{241}Am-Be	-----	-----	1.0	1.0	1.0	3.0
^{241}Am-Be	0.7	300	0.7	1.0	0.7	3.0

* Irradiation distance: distance between the geometrical center of neutron source and sensitive center of rem counter.

Table 3 Calculated response functions of TLD rem counter

Neutron Energy, MeV	Thermoluminecence Intensity per unit Fluence, mR/(n/cm^2)
2.5E-08*	4.5E-06
1.0E-06	8.2E-06
1.0E-05	1.1E-05
1.0E-04	1.5E-05
1.0E-03	1.8E-05
1.0E-02	2.3E-05
2.0E-02	2.5E-05
1.0E-01	3.5E-05
2.0E-01	4.5E-05
5.0E-01	6.9E-05
1.0E+00	9.2E-05
2.0E+00	1.1E-04
5.0E+00	1.0E-04
1.0E+01	7.7E-05
2.0E+01	5.9E-05

* Average energy

Table 4 The response and calibration factor of TLD rem counter to various neutron fields

Neutron source/field	TLD #1 readings				TLD #2 readings				Average neutron Reading, mR	Delivered neutron dose, mSv	Calibration Factor, x10 mSv/mR
	Element 1	Element 2	Element 3	Element 4	Element 1	Element 2	Element 3	Element 4			
^{252}Cf	134mR	121mR	8.9mR	7.5mR	105mR	114mR	7.0mR	7.5mR	111	0.372	0.337
^{252}Cf +PMMA 13.5mm	103mR	103mR	6.6mR	6.8mR	121mR	123mR	5.2mR	4.5mR	107	0.335	0.314
^{252}Cf +PMMA 35.0mm	91.3mR	94.1mR	5.7mR	8.4mR	98.5mR	95.4mR	4.9mR	7.2mR	88	0.262	0.297
^{252}Cf +PMMA 59.0mm	74.2mR	71.6mR	7.2mR	6.2mR	62.1mR	74.5mR	9.6mR	5.1mR	0.64	0.199	0.314
^{252}Cf +PMMA 77.5mm	52.0mR	54.7mR	4.8mR	6.7mR	49.5mR	53.2mR	5.6mR	7.9mR	0.46	0.149	0.324
^{241}Am-Be	100mR	92.2mR	4.8mR	5.2mR	77.3mR	87.1mR	6.7mR	7.5mR	0.83	0.286	0.344

Table 5 Counting responses of the two conventional rem counters to various neutron fields

		^{252}Cf	^{252}Cf +PMMA 13.5mm	^{252}Cf +PMMA 35.0mm	^{252}Cf +PMMA 59.0mm	^{252}Cf +PMMA 77.5mm	^{241}Am-Be at 1.0m	^{241}Am-Be at 0.7m	Background Counts
Delivered Neutron Dose Rate (mSv/h)		0.223	0.201	0.157	0.115	0.090	0.287	0.572	-----
Fuji NSN1	Counts/sec	899	822	650	488	375	97	189	218counts per 10min
	Counts/sec per 10μSv/h	40.2	40.9	41.3	42.4	41.9	33.8	33.0	
Studsvik/Alnor 2202D	Counts/sec [1)]	79	69	53	38	29	8.8	18	34counts per 10min
	Counts/sec per 10μSv/h	3.5	3.4	3.4	3.3	3.3	3.1	3.2	
	Counts/sec [2)]	84	74	57	42	32	10	19	
	Counts/sec per 10μSv/h	3.7	3.7	3.6	3.6	3.6	3.5	3.4	

Incident direction, 1) to the top 2) to the lateral side