APPLICATION OF MCP-N (LiF:Mg,Cu,P) TL DETECTORS IN MONITORING ENVIRONMENTAL RADIATION

by

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Thermoluminescent MCP-N detectors based on LiF:Mg,Cu,P are by about 2 orders of magnitude more sensitive than TLD-100 detectors based on conventional LiF:Mg,Ti, which makes it possible to use them in short-term monitoring of ionising radiation in the environment (*e. g.*, over a two-week period, rather than over 3-12 months). We describe the properties of MCP-N detectors and methods of their application in environmental monitoring. The system was tested in short and long-term exposure periods at 100 sites around Krakow region. MCP-N detectors were then applied to measure variation of radiation dose rate at four selected villages in Serbia, where depleted uranium ammunition was deployed in 1999. Together with short-term thermoluminescent dosimetry, *in situ* measurements using proportional counters were performed in order to assess the range of variation of natural radiation background in these villages. The mean terrestrial kerma dose rate in these villages was found to vary between 85 and 116 nGyh^{-1} and the average ambient dose equivalent rate $H^*(10)$ determined by thermoluminescent detectors and by proportional counter measurements was 160 nSvh^{-1}. These values of natural radiation background dose rates can be applied as reference levels for field measurements around other sites where depleted uranium ammunition was deployed.

Key words: thermoluminiscent detector, lithium fluoride, depleted uranium, MCP-N, environmental dose

INTRODUCTION

Since 1978, when Nakajima first reported on the properties of a new heavily doped thermoluminophor LiF:Mg,Cu,P, the "high-sensitive" lithium fluoride has become a widely applied detector in radiation protection and environmental monitoring. The high sensitivity of this detector permits it to be applied over a broad range of environmental surveys: in short-term measurements, lasting from a few hours to a few days, and in long-term monitoring which lasts for months or years [1, 2]. In 1987, MCP-N (LiF:Mg,Cu,P) sintered pellets, of 4.5 mm diameter and 0.9 mm thickness, were first developed by Niewiadomski [3] at the Institute of Nuclear Physics (INP, Polish acronym IFJ) in Krakow, Poland. Over the following years, in the same laboratory, the properties of these detectors, such as fading, light sensitivity, Lowest Detectable Dose, self-dose, zero-dose, energy response from low-energy X-rays up to 6-7 MeV, and the influence of annealing and readout conditions on detector stability, have been systematically studied and optimised. MCP-type detectors are now commercially available from TLD Poland [4].

Environmental monitoring of radiation doses using passive thermoluminescent (TL) detectors is an extremely valuable companion to environmental monitoring using systems equipped with active, on-line dosimeters. Thermoluminescent detectors

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(TLD) are frequently used for long term (3-12 months) environmental dosimetry, *e. g.* around nuclear installations. The so-called high-sensitive TLD's based on LiF:Mg,Cu,P are about 2 orders of magnitude more sensitive than conventional LiF (TLD-100) which enables short-term (2 weeks, against 3 months exposure) radiometry surveys of the environment [5]. This high sensitivity also allows short-term (hourly or daily) monitoring of local changes of radiation doses after accidental contamination of the environment. Since mid-90's, MCP-N detectors have been routinely used for monitoring of environmental doses around the nuclear facilities of the Institute of Nuclear Physics in Krakow.

An environmental monitoring system that uses MCP-N detectors, and consists of TLD cards (three TLDs per card), automatic TL readers, heating ovens, and specially developed software which includes a data base for the evaluation of results, was next developed and field-tested at the INP. The system was also tested in short- and long-term exposure periods at 100 sites around the Krakow region [6].

We now decided to further test MCP-N detectors against real-time measurements *in situ* using an ionization chamber, to measure the variation of environmental radiation dose rate at selected sites in Serbia, where depleted uranium (DU) ammunition was deployed during the 1999 Balkan conflict. The goal of these measurements was to assess the range of variation of the natural radiation background in villages where DU ammunition was deployed. The measured values of natural radiation background dose rates could then be applied as reference levels for field measurements around other areas where DU ammunition was found.

The aim of this work is to present the results of environmental monitoring of radiation doses in four villages in Serbia where DU ammunition was deployed. We also review the basic properties of MCP-N detectors and describe in some detail the methods of their application in environmental radiation monitoring.

MATERIALS AND METHODS

Properties of MCP-N detectors

The main parameters of MCP-N such as fading, light sensitivity, Lowest Detectable Dose, L_D, self-dose, zero-dose, energy response from low-energy X-rays up to 6-7 MeV, influence of annealing and readout conditions on detector stability, were investigated and optimised [2, 6-12]. The values of these parameters, against the ones that refer to standard LiF:Mg,Ti (MTS-N, TLD-100 equivalent) detectors, are listed in tab. 1.

Table 1. Main parameters of MCP-N detectors

Parameter	Value
Sensitivity (as compared to TLD-100)	25 times higher
Lowest detectable dose, $L_{D,}$ [6, 7]	60 nGy
Linearity range [8]	from 200 nGy to 5 Gy
Self dose [2]	less than 1 nGyh^{-1}
Fading, [9]	less than 5% per year
Sensitivity to light [10]	negligible
Energy response (60 keV to 6-7 MeV) [11]	<20%
Zero-dose [6]	150 nGy
Time estimation using peak ratios [12]	up to 3 months

Procedures and instrumentation

The golden rule for handling LiF:Mg,Cu,P detectors is never to heat them above 240 °C, as this can lead to a decrease of detector sensitivity. Therefore, during readout it is recommended either to apply step heating up to 240 °C or linear heating up to about 260-270 °C, but for a limited time of a few seconds only. The recommended annealing procedure consists of heating for 10 minutes at 240 °C, followed by rapid cooling on a thick aluminum block, at room temperature. TLD reader annealing at 240 °C is also possible. After their irradiation detectors are additionally annealed for 10 minutes at 100 °C to remove the low-temperature peaks.

The instrumentation and standard procedures applied in the environmental monitoring system developed at the Institute of Nuclear Physics (IFJ) are presented in tab. 2. The environmental dosimeter consists of a KD-85 dosimetry card containing three highly sensitive MCP-N (LiF:Mg,Cu,P) detectors closed in a 2 mm thick PCV container, packed in a light-tight aluminum foil and a waterproof polyethylene bag. The package is mounted on a steel rod at the height of 1 m above ground level. For readout, the ACARD97 automatic TL reader (Microlab, Poland) is used. Detectors are annealed in the PTW-TLDO programmable furnace. The environmental dosimeters are calibrated free in air, in terms of kerma in air, using the ^{137}Cs beam at the Laboratory for Calibration of Radiation Protection Instruments at IFJ.

FIELD MEASUREMENTS

Tests of the system

Over the period June-December 1999 the system was tested in approximately 100 field sites over Southern Poland [13, 14]. Two exposure times were chosen: 96 hours (sign "SP" for short-period) and 2160 hours ("LP" for long-period). The preparation of 100 dosimeters for distribution, including

Table 2. Instrumentation and procedures used in the IFJ environmental monitoring system

Element	Instrumentation
TL detectors	LiF:Mg,Cu,P chips (trade name MCP-N)
TL cards	KD96 aluminum cards containing three MCP-N detectors, wrapped in teflon foil
TL readers	Two ACARD97 automatic, microprocessor-controlled ohm-heating readers. Both readers are fully compatible
Annealing ovens	Carbolite 400 and PTW TLD0 microprocessor-controlled ovens
Standard software	ACARD software for controlling the reader and simle TL glow curve and database operations
Annealing	Standard: 10 minutes at 240 °C±1 °C
Distribution	Rapid deployment using transportation by car, with well-known transit-dos
Exposure sites	1 m above ground
Readout	Linear heating up to 300 °C or three-step plateau heating (3 s at 150 °C, 15 s at 260 °C, and 3 s at 260 °C)
Evaluation of results	Integration of glow-curves

heating of the annealing oven to a temperature of 240 C, annealing the detectors, packing them into containers, packing the containers into foil, placing identification stickers on the dosimeters and packing them into a transportation container, takes two technicians about 4 hours. Meanwhile, a table is printed out with detector ID numbers and planned locations of measurement points at which the date and time of detector installation should be recorded. Metal rods, which allow the dosimeters to be exposed at the height of 1 m above ground, are stored in a separate container. The dosimeters are distributed by cars to the measurement sites, which are placed at weather (meteorological) stations at distances up to 150 km from the IFJ, and in private gardens of volunteers in the Krakow city area. It took approximately 12 hours to distribute 100 dosimeters (35 outside Krakow, 35 within the city of Krakow and 30 around the IFJ laboratory area). After 4 days or 3 months of exposure the dosimeters were collected and read out 6 hours within 12 hours after their collection.

The results obtained during these test measurements demonstrated that the kerma rates measured over a 4-day screening period coincide well with kerma rates determined during a 75-day monitoring period. The response of LiF:Mg,Cu,P is strongly dependent on radiation density (LET). If detector calibration is performed with ^{137}Cs or ^{60}Co -rays, low-LET muons (present in the cosmic-radiation component) and high-energy -rays from the thorium and uranium series, are registered with higher efficiency in LiF:Mg,Cu,P than in LiF:Mg,Ti. Therefore, the kerma rates obtained during 75-day measurements using conventional LiF:Mg,Ti (MTS-N) detectors were found to be 10-20% lower than those obtained with MCN-P detectors, due to the different energy response of LiF:Mg,Ti and LiF:Mg,Cu,P materials. The measured kerma rates at meteorological stations measured in 1999-2000 coincide within 5-15% with values measured at the same stations in mid 80's, before the Chernobyl accident [14].

Survey of environmental doses at selected area in Serbia

Dosimeter packages with MCP-N detectors were used to measure environmental radiation doses in four villages of Serbia to determine the variation of environmental dose rates in villages where the DU ammunition had been deployed during the 1999 Balkan conflict. A hundred packages were prepared at IFJ, sent by courier post and distributed by the VINČA Institute team in Bratoselce, Borovac, Pljačkovica, and Reljan. The dosimeters were exposed in open air, mounted on metal rods 1 m above ground level. Detectors were collected after a 3-month exposure period and sent for evaluation to IFJ. The transport dose before the collection and readout was estimated by transport detectors.

The kerma-rate from terrestrial radiation was calculated by subtracting the calculated values of cosmic-ray kerma-rates at the given altitude from the measured values of kerma-rate. The mean values of air kerma-rate due to cosmic-ray exposure at the sea level determined in the past [1, 2] for MCP-N (32.1 nGyh^{-1}) corresponds well with the recently obtained [14] value of 32 nGy/h. The cosmic ray dose rate, as calculated with the CARI-LF code for solar potential for spring 2002 and the co-ordinates of Borovac (altitude 42.3 N, longitude 21.4 W) increased from 29.6 nGyh^{-1} at sea level to 33 nGyh^{-1} at the altitude of 520 m, *i. e.*, by 11.5%. Therefore for Reljan, Bratoselce, and Borovac the response of MCP-N detectors was estimated at 1.115 32.1 nGyh^{-1} = 35.7 nGyh^{-1}. The cosmic-ray dose rate for Pljačkovica (altitude approximately 1100 m) calculated with CARI-LF was 37.8 nGyh^{-1} which leads to the estimated response of MCP-N: 37.8/29.6 32.1 = 40.9 nGyh^{-1}.

The distribution of kerma-rate in air, measured with MCP-N detectors in Pljačkovica, Reljan,

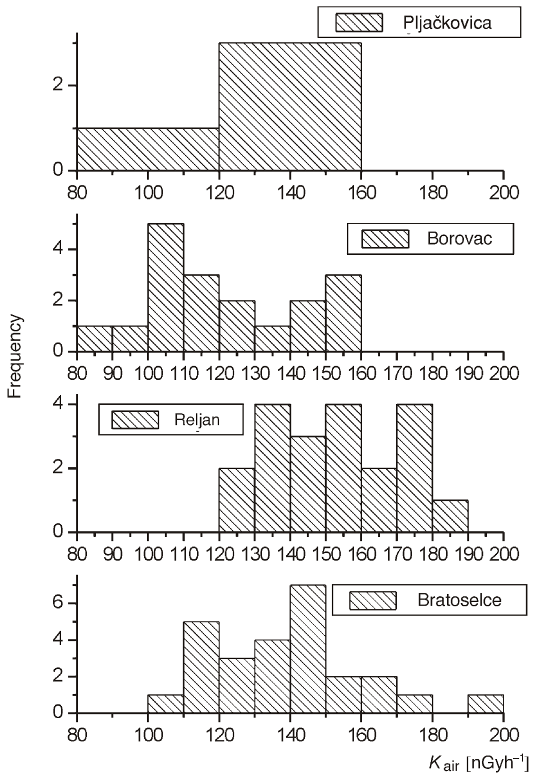

Figure 1. The distribution of values of kerma rates in air, measured in Pljačkovica, Borovac, Reljan and Bratoselce in the spring of 2002 using dosimeters with MCP-N (LiF:Mg,Cu,P) TLD

Bratoselce, and Borovac in the spring of 2002, are presented in fig. 1. Total kerma in air was determined in the measurements *i. e.* kerma in air originating both from terrestrial and cosmic-ray radiation. The variation of the kerma-rate at different measurement sites is significant (range 80 to 200 nGyh^{-1}), and depends on the type of bedding at the measurement site.

In tab. 3 the mean values of total kerma-rate and the mean value of terrestrial kerma-rates at the selected villages in Serbia in spring 2002 are presented. The terrestrial kerma-rate was calculated by subtracting the contribution of cosmic-rays (35.7 nGyh^{-1} for Reljan, Bratoselce, and Borovac and 40.9 nGyh^{-1} for Pjačkovica) from the total kerma-rate. The values of

Table 3. Mean kerma rate measured with MCP-N detectors at selected villages in Serbia in the spring 2002. The terrestrial kerma rate was calculated by subtracting form the total the estimated dose rate due to cosmic rays

Site	Mean K_{air} total [nGyh^{-1}]	Mean K_{air} terrestrial [nGyh^{-1}]
Bratoselce	138 21	102 22
Reljan	152 ± 18	116 ± 19
Borovac	121 21	85 ± 22
Pljačkovica	133 ± 29	92 ± 30

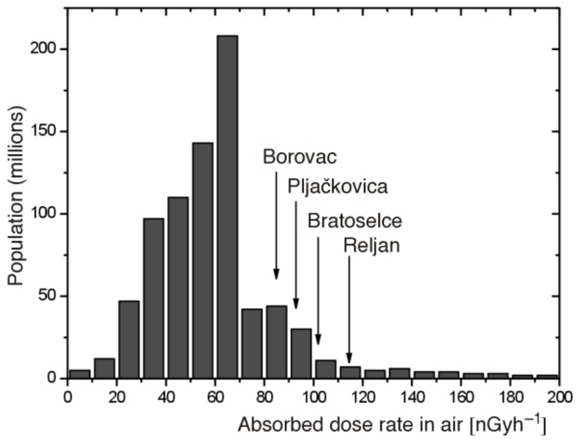

Figure 2. Comparison of mean values of kerma-rates in air from terrestrial radiation, determined in Borovac, Pljačkovica, Bratoselce, and Reljan in the spring of 2002 using MCP-N (LiF:Mg,Cu,P) TLD, with the distribution of absorbed dose in air, weighted over the exposed population, as given by UNSCEAR [15]

the mean terrestrial kerma-rate in all villages (between 85 and 116 nGyh^{-1}) bring the population in these villages to the upper 20% of natural radiation dose rates in the world [15] (see fig. 2).

In 52 selected sites where TLDs were installed, additional *in-situ* measurements with a proportional counter were performed by the group from the Jožef Stefan Institute, Slovenia. The proportional counter was calibrated in terms of ambient dose equivalent, $H^*(10)$. The values of $H^*(10)$ for TLDs were calculated by applying the conversion coefficient $H^*(10)/K_{air}$ = 1.20 Sv/Gy for ^{137}Cs -rays. The values of $H^*(10)$ determined using both methods are compared in fig. 3. and in tab. 4. Correspondence between the two methods is quite remarkable, considering that

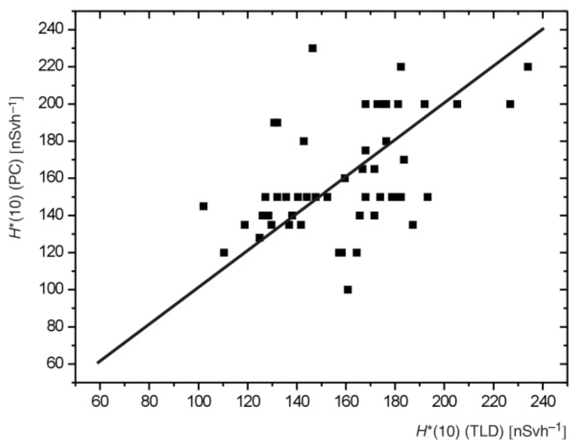

Figure 3. Ambient dose equivalent, $H^*(10)$ determined in 52 selected sites measured at Borovac, Pljačkovica, Bratoselce, and Reljan in the spring of 2002 with a proportional counter (PC) and using MCP-N TDL

Table 4. Comparison of average ambient dose equivalent rates measured at 52 selected sites with the proportional counter (Jožef Stefan Institute, Slovenia) and MCP-N thermoluminescent detectors

	K_{air} [$nGyh^{-1}$]	$H^*(10)$ [$nSvh^{-1}$]
MCP-N (LiF:Mg,Cu,P)	133.1	159.6
Proportional counter	–	160.1

calibration of the two systems (TLDs and proportional counter) was performed quite independently.

CONCLUSIONS

High sensitive MCP-N (LiF:Mg,Cu,P) thermoluminescent detectors were applied in a system of environmental monitoring of radiation doses. Due to the high sensitivity of MCP-N detectors the system allows for short term (hours and days) and long term (months and years) surveys of environmental doses. Due to the low cost of individual dosimeters, it is possible to simultaneously monitor environmental radiation kerma-rates at a large number of locations over a period from a few days to a year, or so. Provided prior reference values of natural background kerma-rates at selected monitoring points are available, the system can be applied to rapidly assess kerma-rates in the environment in case of radiation accidents or radioactive contamination of the environment. The system was tested in short-term and long-term measurements in the Krakow region and in Serbia, in villages where DU ammunition was found. A more systematic survey of environmental dose rates in Serbia will be performed with MCP-N detectors in the years 2004-2005.

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Павел ОЛКО, Маћеј БУДЗАНОВСКИ, Павел БИЛСКИ, Снежана МИЛОШЕВИЋ, Барбара ОБРИК, Ева ОХАБ, Мирослав СИМИЋ, Петер СТЕГНАР, Михал П. Р. ВАЛИГОРСКИ, Зора С. ЖУНИЋ

ПРИМЕНА MCP-N (Lif:Mg,Cu,P) ТЕРМОЛУМИНИСЦЕНТНИХ ДЕТЕКТОРА У МОНИТОРИНГУ ЈОНИЗУЈУЋИХ ЗРАЧЕЊА ИЗ ПРИРОДЕ

Термолуминисцентни MCP-N детектори на бази LiF:Mg,Cu,P су за два реда величине осетљивији од конвенционалних детектора на бази LiF:Mg,Ti, због чега се употребљавају у краткотрајном и оријентационом мониторингу јонизујућих зрачења из природе (што практично обухвата период од преко две недеље уместо 3-12 месеци). У раду се описују својства MCP-N детектора и методе за њихову примену у мониторингу јонизујућих зрачења из природе. Систем је превасходно тестиран у краткотрајним и дуготрајним излагањима јонизујућим зрачењима из природе на преко 100 мерних места у подручју око Кракова. MCP-N детектори су затим употребљени за референтна мерења брзине дозе у околини четири руралне заједнице у Србији, наменски одабране, у којима је 1999. године распршена муниција са осиромашеним уранијумом. Истовремено је приликом постављања ових MCP-N детектора у руралним заједницама, *in situ,* паралелно извршено мерење брзине дозе са пропорционалним бројачем, са циљем процене опсега варијације основног нивоа зрачења из природе у испитиваним руралним заједницама. Средња вредност терестријалне брзине дозе керме у испитиваним руралним заједницама варирала је у опсегу од 85 до 116 nGyh^{-1} а средња вредност амбијенталне еквивалентне брзине дозе $H^{*}(10)$ одређене помоћу термолуминисцентних детектора и пропорционалним бројачем износила је 160 nSvh^{-1}. Ове измерене вредности основних нивоа зрачења из природе могу се користити као референтни нивои теренских мерења у околини свих других подручја погођених муницијом са осиромашеним уранијумом.