

Optically Stimulated Luminescence Materials for Wide-spectrum Neutron Measurements at CERN

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Abstract

For the first time, Optically Stimulated Luminescence (OSL) materials with enhanced neutron-sensitivity have been irradiated. The neutron irradiations were carried out in the accelerator-like environment of the IRRAD2 facility at CERN, Geneva, Switzerland and in the TRIGA reactor at JSI, Ljubljana, Slovenia. The results show the possibility to measure with high accuracy the thermal and the fast components of these two different neutron spectra by means of OSL materials doped with Boron or mixed with Polyethylene. The applicability of this technology in the measurement of the neutron components in the complex radiation environment expected at CERN LHC experiments is therefore demonstrated.

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I. INTRODUCTION

THE radiation environment expected in the future CERN Large Hadron Collider (LHC) experiments will be composed of neutrons, photons and charged hadrons. This complex field, which has been simulated by Monte Carlo codes [1, 2], is due to particles generated by the proton-proton collisions and reaction products of these particles with the sub-detector materials of the experiment itself. Neutrons, becoming dominant at large radii with respect to the interaction point, are a matter of concern due to radiation damage to all exposed particle detectors and electronic components [3]. For example, in the EndCap electromagnetic calorimeter of CMS as well as in the innermost barrel layer of the ATLAS liquid argon calorimeter, an annual 1 MeV equivalent neutron fluence of more than 10^{13} cm^{-2} is expected.

An integrated sensor based on different dosimetric technologies, including Optically Stimulated Luminescence (OSL) materials, RadFETs and $p-i-n$ diodes is currently under development for the radiation monitoring of the LHC experiments [4]. An overview of this work was already given elsewhere [5, 6]. The present paper details a mandatory step in the OSL qualification process: the evaluation of neutron sensitivity and neutron energy dependence.

OSL materials as radiation dosimeters for medical, space and High-Energy Physics (HEP) applications have been studied extensively [7, 8]. For HEP applications, successful experiments were carried out to measure the dose delivered by different charged particles [9]. The OSL response to neutron irradiation has still to be studied, as a prerequisite to a possible use in the CERN radiation environments.

A first irradiation campaign was thus carried out on special OSL materials with an enhanced sensitivity to different neutron energies. The OSL standard materials were doped with Boron or mixed with polyethylene to increase their sensitivity to thermal and fast neutrons respectively. The samples, prepared from the *Electronic and Radiations Research Laboratory* of the University Montpellier II, France, were irradiated in the IRRAD2 Facility at the CERN Proton-Synchrotron (PS) [10] and at the TRIGA reactor facility at the *Jozef Stefan Institute (JSI)* of Ljubljana, Slovenia [11].

The principles of OSL dosimeters used for neutron detection are reviewed in *section II*. In *section III*, the experiments in the two neutron facilities are briefly described. In *section IV*, the results are summarized and discussed. Finally, our conclusions are given in *section V*.

II. REVIEW OF OSL MATERIAL AS NEUTRON DETECTOR

The OSL materials have the property to emit visible light proportional to the received dose of radiation.

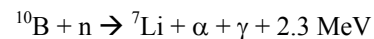
Ionizing radiation creates electron-hole pairs in the OSL material. A fraction of these carriers is trapped in energy levels located in the wide band gap of the insulator. These

charges will remain trapped for a period of time depending on the activation energy of the traps and the temperature. When the energy necessary to release the charges is provided by an optical stimulation, a radiative recombination may be observed. Quantifying the amount of the emitted light makes it possible to evaluate the dose [12].

Neutrons do not produce direct ionization in materials; however, a little amount of ionizing energy is deposited via secondary particles generated by neutron interactions. This energy deposition is usually not sufficient to generate a measurable quantity of electron-hole pairs in pure OSL.

For this reason, different additives were mixed with the standard OSL material that consists of doped Strontium Sulfide (SrS). In particular: Boron (B) was added to increase the OSL sensitivity to thermal neutrons and polyethylene (PE) was added for fast neutron detection.

Boron is responsible for thermal neutron capture via the nuclear reaction:



This reaction has a very large cross section $\sigma_r = 3840$ barns for thermal energies that decreases down to a few barns for energies above some tens of keV as shown in Fig. 1 [13]. The alpha particles and the lithium ions generated by the neutron capture lose their energy via ionization in the sensitive SrS material. The ranges of the two reaction products in the SrS were calculated with the SRIM software [14]. Taking into account that the 2.3 MeV released in the reaction contributes to the kinetic energy of the lithium ion (0.83 MeV) and the alpha particle (1.47 MeV), ranges of $2 \mu\text{m}$ and $4.2 \mu\text{m}$ in SrS were calculated respectively. These short path-lengths guarantee that all captured thermal neutrons deposit the full ionization energy in the OSL material.

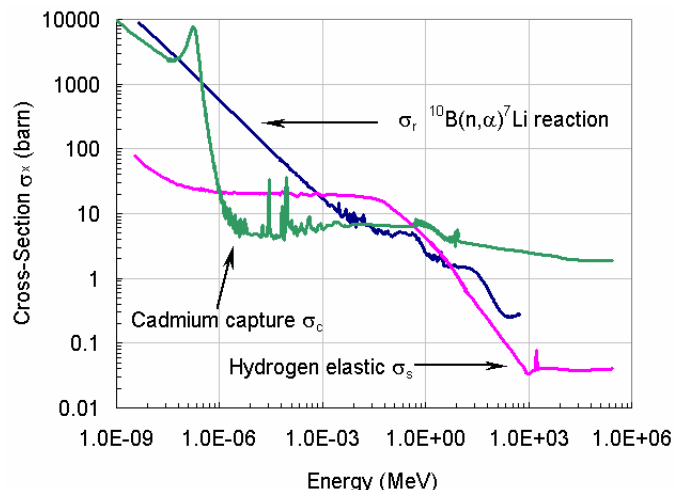


Fig. 1 Plot of the neutron reaction cross-sections of interest for this work [13].

PE with its high content of hydrogen is the preferred material to detect fast neutrons because a colliding particle can transfer from none to all its energy to the proton in one

elastic collision. The average recoil energy will be half of the initial neutron energy. Hydrogen has a cross-section σ_s of the order of tens of barns for epi-thermal neutrons that falls to 4 barns for the energy of 1 MeV. It further decreases to approximately 1 barn when the neutron energy reaches 8 MeV as shown in Fig. 1. The recoil proton will move through the sensitive material with a path length depending on the energy transferred to it by the incident neutron.

III. EXPERIMENTS

III.1. Samples manufacturing

The pure OSL material and three compounds of OSL material doped with Boron and mixed with polyethylene were prepared and irradiated to different neutron fluences.

The four different types of OSL samples are shown in Fig. 2. They have a size of 5x5 mm² and various thicknesses. In particular, the used materials had the following characteristics:

- 1) OSL standard material (*OSL pure*): specimens obtained from a 100 μm thick layer of SrS silk-screen printed on a Kapton foil.
- 2) OSL doped with Boron (*OSL+B*): specimens obtained from a 100 μm thick layer of SrS, 1 %-weight Boron doped, silk-screen printed on a Kapton foil.
- 3) OSL mixed with Polyethylene (*OSL+PE*): specimens gained from a bigger block in which the SrS was melted with polyethylene. The thicknesses of the samples were of a few mm.
- 4) OSL mixed with PE and Boron (*OSL+PE+B*): OSL specimens containing both, PE and Boron. The thicknesses of these samples were also of a few mm.

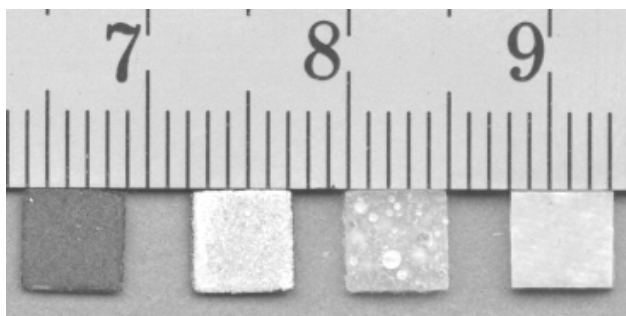


Fig. 2 OSL materials from left to right: pure OSL material, OSL doped with Boron, OSL mixed with Polyethylene and OSL mixed with Polyethylene and Boron.

Of each type of OSL material, 3 to 5 pieces were used in order to prove the reproducibility of the results. They were packaged and irradiated in few micron-thick aluminum foils in order to protect them from light. Before irradiation, the samples were heated to 80 °C for 24 hours to anneal the charge in the OSL trap levels (i.e. to clear the dosimetric material).

III.2. CERN IRRAD2 neutron facility

The irradiation in the facility IRRAD2 at the CERN-PS accelerator is performed in a cavity with secondary particles produced by the primary 24 GeV/c proton beam after crossing a thick beam dump consisting of carbon and iron blocks [10].

A motorized shuttle system allows the transport of samples from the counting room to the irradiation cavity in which a broad spectrum of neutrons is produced as shown in Fig. 3 [15].

Together with neutrons, high-energy hadrons and gamma rays are produced. Gamma rays represent a contamination that is in the order of 10-20 % with respect to the neutron component, depending on the operation conditions.

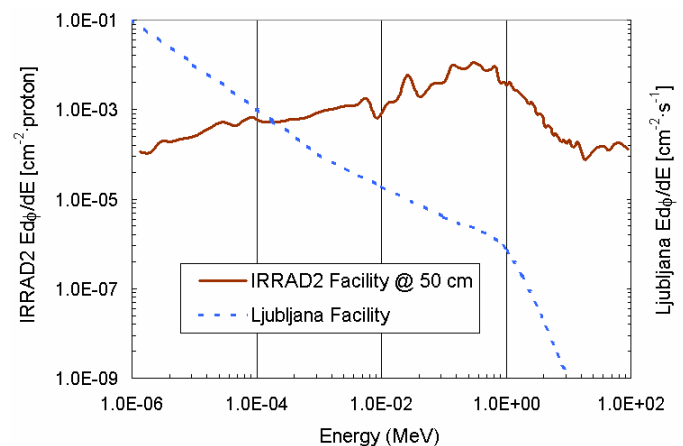


Fig. 3 Neutron spectrum in the IRRAD2 Facility at 50 cm from beam axis normalized to one impinging proton (continuous line). Ljubljana Reactor neutron spectrum as measured from activation foils (dotted line).

With the remote shuttle system, it is possible to set the position of the samples with respect to the beam axis. At positions far from the beam axis, the ratio of charged hadrons in the GeV energy range to neutrons can be considerably reduced and it is thus possible to perform irradiations in an almost pure neutron environment. The OSL samples were thus placed at 50 cm from beam axis and exposed to four different 1 MeV neutron equivalent fluences, reaching a maximum value of $1.6 \times 10^{12} \text{ cm}^{-2}$.

III.3. TRIGA reactor Ljubljana neutron facility

The Reactor Research Centre at the *Jozef Stefan Institute (JSI)* in Ljubljana has an experimental nuclear reactor of type TRIGA [11].

The neutron spectrum of this facility, determined by measuring the activation of foils of different materials, is shown in Fig. 3.

Irradiations were performed placing the OSL samples into the reactor core through an irradiation tube that occupies a fuel rod position and enters the reactor core from above. The neutron flux delivered to the samples is linearly proportional to the reactor operating power.

A first series of OSLs was placed in the core, and exposed to three different 1 MeV neutron equivalent fluences, reaching a maximum value of $6.9 \times 10^{11} \text{ cm}^{-2}$. A second series was shielded by means of a 1.0 mm thick Cadmium (Cd) foil in order to irradiate the OSLs without the thermal neutron component of the reactor spectrum (see σ_c behavior in Fig. 1). The Cd thickness was chosen according to the thermal neutron mean-free-path that is of about 100 μm .

III.4. Dosimetric measurements during irradiations

Gafchromic[®] Dosimetric Films HD-810 [16] and silicon detectors [17] were used in both facilities to measure the Total Ionizing Dose (TID) and the neutron fluence respectively.

The film Gafchromic[®] HD-810 develops a characteristic blue color upon exposure to ionizing radiation and becomes progressively darker in proportion to the absorbed dose. The color change has been measured after irradiation on samples of 1 cm^2 by means of a transmission densitometer. The film calibration used in the present work was performed using the 24 GeV/c protons of the IRRAD1 facility at CERN [18].

Silicon detectors are reverse biased *p-i-n* diodes with a very high resistivity n-type bulk. Neutron irradiation of such devices will produce generation/recombination centers in the silicon bulk leading to a fluence proportional increase of the leakage current when the device is kept fully depleted [19]. With this technique, it is possible to obtain fluence measurements with an accuracy of $\pm 10\%$.

In the CERN facility, silicon detectors and films were exposed together with the OSLs, while only films were used during Ljubljana irradiations, since the neutron flux in the reactor was previously calibrated.

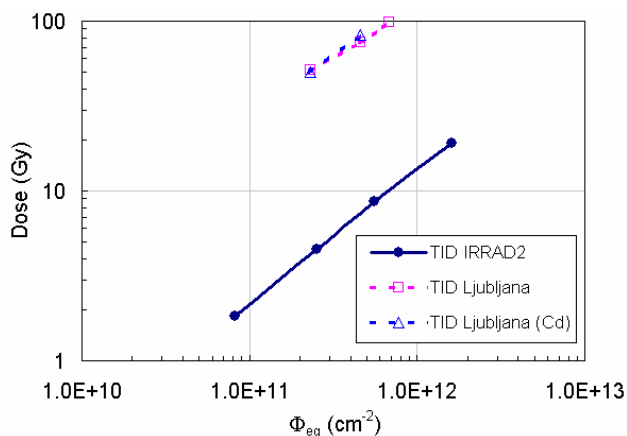


Fig. 4 TID measured in both irradiation facilities by means of Gafchromic[®] sensitive film HD-810.

Fig. 4 shows the results of the TID measurements performed in the two facilities. In the Ljubljana reactor, the TID measured for the samples irradiated in a Cd shielding (triangular markers) is presented separately from the unshielded samples (square markers).

Due to the high gamma background in the Ljubljana reactor, the TID results are about a factor 10 higher than in the CERN facility.

IV. RESULTS & DISCUSSION

All samples were stored at room temperature and measured a few weeks after irradiation. From previous experiences carried out on pure SrS material, the relatively weak fading of OSLs was expected to be stabilized at the readout time [20]. The OSL reader used in this work is an infrared LED focused on the irradiated specimen that is positioned in a samples holder in a light-tight box. The emitted luminescent signal is collected by means of a photomultiplier, then digitalized and recorded by a PC connected to the test bench via a RS-232 interface [7, 9].

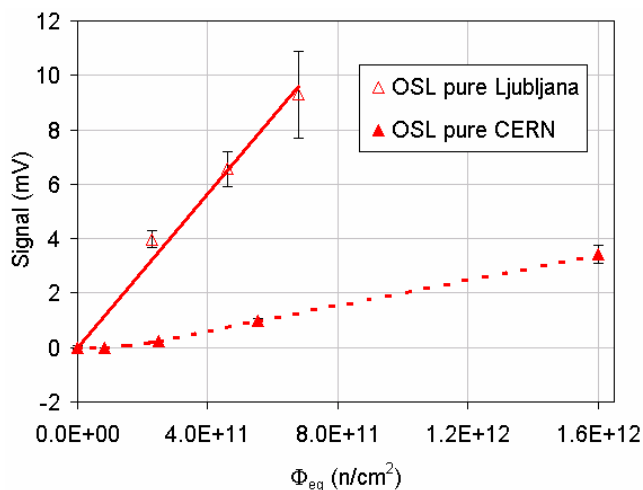


Fig. 5 Pure OSL material response after irradiation at CERN (filled markers) and Ljubljana (empty markers).

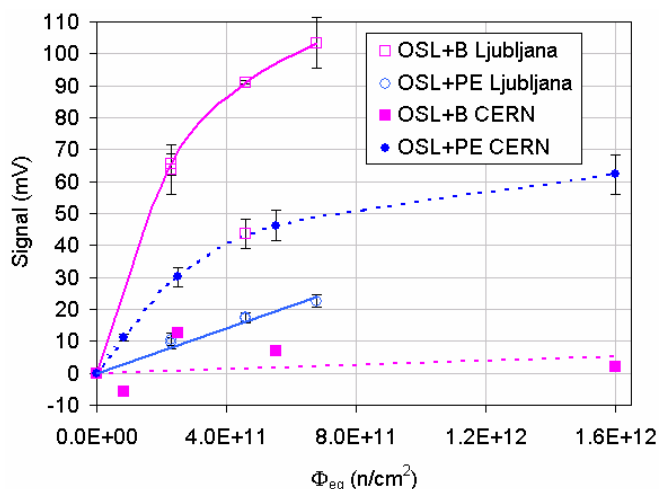


Fig. 6 OSL+B and OSL+PE material response after irradiation at CERN (filled markers) and Ljubljana (empty markers).

The signal (voltage) measured with the above described test-bench has been normalized to the pure OSL sample's

weight in order to take into account the different sample's volume. The so treated signals expressed in mV have been plotted versus the 1 MeV neutron equivalent fluence, and are presented in Fig. 5 and Fig. 6.

After both irradiations, the signal achieved with the *OSL pure* material is the smallest recorded in our tests, and is less than 10 mV as shown in Fig. 5. In the CERN facility (triangular filled markers), for values of 1 MeV neutron equivalent fluence below $1.0 \times 10^{11} \text{ cm}^{-2}$, the signal was lower than the noise level since the gain of the read out system was set to measure high doses. For the Ljubljana facility (triangular empty markers) the signal is a factor 10 higher with respect to the one measured at CERN. This is in good agreement with the TID measurements presented in Fig. 4 of section III.4. As expected, the *OSL pure* material measured thus the ionizing component of the field and is essentially insensitive to neutrons.

TABLE I
RELATIVE SENSITIVITY OF OSLs TO NEUTRONS

	OSL+B / OSL pure	OSL+PE / OSL pure
CERN	1.6	22.0
Ljubljana	20.1	2.5

Fig. 6 shows the results obtained for the *OSL+B* and *OSL+PE* materials.

The signal measured with these two neutron-sensitive materials shows up to a factor 20 increased sensitivity with respect to the *OSL pure* samples as shown in Table 1.

In the Ljubljana facility (continuous lines in Fig. 6), the measured signals from the *OSL+B* (square empty markers) and *OSL+PE* (round empty markers) samples represent 87 % and 13 %, respectively, of the total neutron signal. At CERN (dotted lines in Fig. 6), the signals obtained from the *OSL+PE* samples (round filled markers) represent more than 97 % of the neutrons signal, as the *OSL+B* (square solid markers) samples gave only a negligible signal.

All the above results are in very good agreement with the spectra composition of the two neutron environments as shown in Fig. 3 of section III.2 and summarized in Table 2.

TABLE II
SPECTRA COMPOSITION IN THE NEUTRON FACILITIES

	Thermal (< 1eV)	Epi-Thermal (1eV-50keV)	Fast (50keV-1MeV)	High-Energy (>1MeV)
CERN	0.1 %	15.3 %	63.5 %	21.1 %
Ljubljana	89.0 %	10.9 %	< 0.1 %	< 0.1 %

The strongly fluctuating signal of the *OSL+B* samples obtained at the CERN facility can be explained as a difficulty in the measurement of a signal that is close to the electronic noise level of the readout apparatus.

Moreover, one of the samples irradiated in Ljubljana also shows a reduced response in the same fluence range (see the square empty markers at $4.6 \times 10^{11} \text{ cm}^{-2}$ of Fig. 6). Again, an

experimental problem is suspected, which requires crosschecking in a forthcoming irradiation. However, at this moment it is not possible to exclude a possible influence of the Boron doping on the OSL's annealing behaviour.

For the *OSL+PE+B* only the Ljubljana data are available. This material gave the same response as the *OSL+PE* that is already plotted in Fig. 6. The current method to produce *OSL+PE+B* specimens leads to a predominant polyethylene fraction compared to the Boron one and is likely to account for this effect.

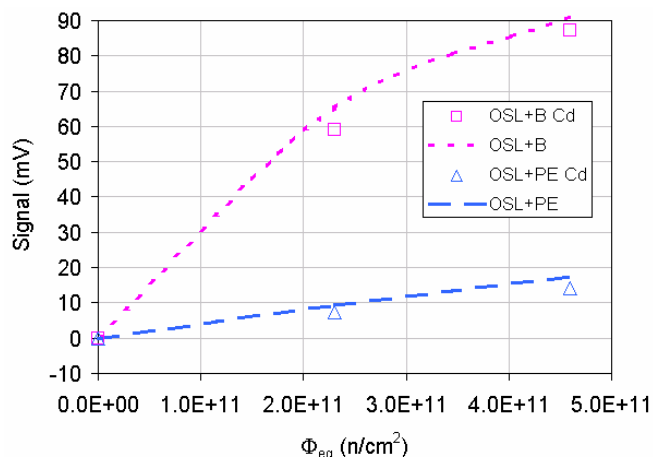


Fig. 7 Series of samples irradiated in the Ljubljana reactor while a Cd shielding suppressed the thermal neutron component of the spectrum (markers). The signals measured for the samples irradiated with the whole spectrum (unshielded samples) are also shown for comparison (dashed lines).

Finally, for the Ljubljana samples, an inter-comparison of *OSL+B* and *OSL+PE* samples irradiated with and without the thermal neutron component of the spectrum (i.e. without or with Cd shielding) is presented in Fig. 7.

The shielded *OSL+B* and *OSL+PE* samples show a 10 % reduced response. However, the signal coming from the Cd-shielded *OSL+B* samples was expected to be significantly reduced with respect to the unshielded ones. This does not occur and is probably because of the rapidly falling Cd cross-section for neutron captures (σ_c) in the epi-thermal energy range. It decreases much faster than the Boron capture cross section (σ_b) as visible in Fig. 1.

The relative error evaluated on voltage measurement over all series of irradiated OSL materials was 7.9 % and 12.0 % for the Ljubljana and for the CERN samples respectively, proving a good reproducibility of the measured results. For doses exceeding the mGy range, the precision achievable with the OSL technology is however of about 3 % [20].

The systematic error that influences the presented measurements it is probably due to misalignments that can occur during the positioning of the irradiated specimens in the samples holder of the test-bench. Therefore, the reduction of this error source has been taken into account in the manufacture of the new OSL samples presented in the following section.

V. CONCLUSION & OUTLOOK

For the first time the different components of wide neutron spectra have been measured by means of specially prepared Optically Stimulated Luminescence materials (OSLs).

Results of neutron irradiations carried out in the accelerator-like environment of the CERN-PS IRRAD2 facility and in the environment of the TRIGA reactor in Ljubljana were presented. Standard OSL material (SrS) doped with Boron or mixed with polyethylene allowed to measure with high accuracy the thermal and the fast components of these two completely different neutron spectra.

However, further tests have to be performed to better understand the behaviour of the samples doped with Boron, and any possible influence of the doping on the material's annealing behaviour. The influence of the sample's thickness on the response will also be investigated in detail.

These results already prove that properly prepared OSLs are a promising option for the monitoring of the different neutron components in the complex radiation environments of the CERN LHC experiments.

New specimens of pure and neutron-sensitive OSLs have already been prepared. To improve the samples readout, the *OSL pure* and *OSL+B* materials have been deposited directly on the active surface of GaAsP visible photodiodes sealed in a TO-5 package. For fast-neutron detection the package of some *OSL pure* samples were then filled up with paraffin.

Further neutron irradiations over an extended fluence range will be performed. With these samples the feasibility of a "real-time" readout, as desirable for radiation monitoring in the CERN LHC experiments, will also be tested.

VI. ACKNOWLEDGMENTS

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