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Fiber Optic remote gamma dosimeters based on Optically Stimulated Luminescence: State-of-the-art at CEA

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Abstract. Optically Stimulated Luminescence (OSL) used in conjunction with optical fibres allows remote measurements of both dose and dose rate for the purpose of radiation protection in nuclear industry and medecine (radiology, radiotherapy), monitoring of installations (dismantling) as well as in process controls. Since 1995, two types of OSL systems have been developed by the Optical Measurement Laboratory (CEA) for gamma detection. The first system relies on rare-earth-doped Alkaline-Earth Sulphides and operates in the red/near-infrared part of the spectrum. It does not provide a tissue-equivalent dose measurement and is dedicated for dismantling operations and process control where a great fiber length is required (100 meters). Several systems of this kind are routinely used by COGEMA and CEA for assistance in their dismantling operations as they provide real-time *in-situ* operation in areas out of reach for usual dosimeters. The second system relies on alumina crystals and operates in the blue/green part of the visible spectrum. It has been developed for radiation protection purposes and may involve a patented compensation technique (both for angular incidence and photon energy).

1. Introduction

Real-time remote dose measurements may be very useful in many applications such as radiation Protection of workers, recycling and dismantling operations, long-term nuclear storage, industrial process controls (*e.g.* X- or e-beam polymerization) and medecine (radiology, radiotherapy).

Optically Stimulated Luminescence (OSL) has been found very useful in all these applications because the dose and dose rate data can be remotely obtained *via* an optical fiber link. This paper describes the state-of-the-art of this technique at CEA-List through industrial applications in the field of dismantling and radiation Protection of workers of the nuclear industry. Upcoming medical applications (radiotherapy, radiology) will be also described in the framework of the MAESTRO European Integrated Project (*Methods and Advanced Equipments for the Simulation and Treatment in Radio Oncology*).

2. Principle of Optically Stimulation Luminescence (OSL)

Optically Stimulated Luminescence is closely related to Thermoluminescence. As for ThermoLuminescent Dosimeters (TLDs), free electrons induced by ionising radiation are trapped on stable energy levels in OSL materials. Then, instead of being released thermally, electrons are released on request, by stimulation light, and produce a luminescence signal which decays as the trap levels are emptied. The integral (over time) of this optical signal is proportional to the dose absorbed by the material between each stimulation. An average dose rate is then estimated taking into account the time elapsed between two successive stimulations. The time decay of OSL is inversely proportional to light intensity (W.m⁻²) and depends also on the stimulation spectrum of the material and the laser wavelength. The reset time may change considerably depending on the laser used for stimulation.

OSL dosimeters offer the same advantages as TLDs (linearity, tissue-equivalence) with the interesting possibility of a remote optical stimulation that enables *in-situ* real-time operation as the reset time of the material ranges from some tenths to tens of seconds. Similarly to TL, OSL provides safety in the advent of an electronic breakdown because the measurement of the dose stored in the material can be delayed as long as necessary.

The OSL phenomenon involves trap centers (labeled as co-activators) and recombination centers (called activators) with energy levels situated into the crystal bandgap (fig. 1). Following an optical stimulation, trapped electrons are freed to the conduction band. These electrons then recombine with so-called recombination centers emitting shorter wavelength photons. The fact that the energy

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(respectively the wavelength) of OSL photons is higher (respectively shorter) than stimulating photons is a typical signature of the OSL effect. This fact - improperly called anti-Stokes luminescence as it is emitted in a spectral domain apart from traditional Stokes fluorescence – enables very small amount of luminescence to be detected (photon counting) even in the presence of laser light because it does not create additional background noise.



Several OSL materials have been studied in the purpose of radiation protection dosimetry in terms of sensitivity and tissue-equivalence. An energy and angular compensation method is generally required to achieve this goal [2]. Rare-earth-doped Alkaline-Earth Sulfides (AES) [3], such as MgS or BaS, are known to be efficient OSL materials but have higher effective atomic numbers (Z > 14) than tissues (Z ~ 7) making compensation very difficult to achieve. Moreover, these materials exhibit an important signal fading at room temperature that requires a calculation to take into account elapsed time and temperature. Finally, these materials are obtained as powder and are opaque to infrared and visible light. Therefore, only a small part of the information can be extracted from the superficial area of the material and collected by the optical fibre. Taking into account these limitations, transparent low-Z OSL materials were investigated in the purpose of increasing the interaction volume and thus the detectivity. The material studied (both of Z=10) were copper-doped silica fibres [4], and alumina (α -Al₂O₃) crystals [5]. Only alumina crystals and silica fibres can be retained as they are characterised by an effective atomic number Z relatively close to tissues, making a compensation scheme feasible. Finally, these non-hygroscopic materials are commercially available at affordable cost.

3. OSL dosimeters developed at CEA

Two generations of OSL systems have been developed at CEA-Saclay in the purpose of a remote OSL dosimetry *via* an optical fibre link.

3.1 First generation system (OSL1)

The first generation system relies on rare-earth-doped AES provided by the University of Montpellier, France [3]. Its operating principle is depicted in fig. 2a [6]. Since it operates in the Near Infra-Red (NIR) optical spectrum, the absorption in silica fibres is typically 10 dB/km which enables remote operation up to several hundreds meters. Since it does not provide a tissue-equivalent indication and enables a dose measurement from 1 mGy up to 10 Gy, it is ideally suited to dismantling and process control operations. For these applications, the sensor is based on a SMA housing ($\emptyset = 5 \text{ mm}$ and 20 mm long) connected to a 200 µm-diameter optical fiber protected in a steel cylinder. A cerium-europium doped barium sulfide polycrystal was coupled to the end of the optical fiber by a connector housing. The optical stimulation was provided by a continuous-wave laser diode (@ 862 nm, 35 mW) and the OSL light was detected by a photomultiplier *via* an optical coupler. This system is now integrated into an industrial PC (fig. 2b).

3.2 Second generation system (OSL2)

The second generation system relies on alumina crystals [4] and operates in the blue-green region of the visible spectrum. In this spectral domain, the absorption in silica fibres is about several 100 dB/km and the practical fibre length is several tens of meters. The OSL2 system has been developed for radioprotection applications that are more stringent because they require a dose resolution of several μ Gy and may involve a compensation technique to allow the sensor measurement to fit the tissue response. The operating principle of the second-generation dosimeter is depicted in fig. 3a. Several crystal pellets are stacked in a sensor housing joined to the fibre connector.

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FIG. 2 : First generation of OSL optoelectronic FIG. 3 : Second generation of OSL dosimeters (OSL 1) a/OSL1: Scheme of operation b/ View of the PC-based system and fiber-connected b/ View of the prototype connected to an OSL transducer

optoelectronic dosimeter (OSL 2) *a*/*OSL* 2 : *Scheme of operation* external PC

In this second generation system (OSL2), the sensor is linked to the optical acquisition unit with an optical fiber (\emptyset 1 mm) of high numerical aperture (0,48). The sensor is 25 mm long and its diameter is 10 mm. Light stimulation is provided by a Diode-Pumped Solid-State (DPSS) YAG-Nd doubled laser (@ 532 nm, 150 mW). Similarly to OSL1, optical filters are inserted into the optical path to remove excess laser light forward (before injection into the fibre) and backward (OSL collection) and a photomultiplier is used for detecting the OSL (fig. 3b). However, several additional equipments were provided in the unit. First, an electromagnetic shutter triggers the stimulation and enables the laser to keep running. Second, an optical collection is provided by an off-axis paraboloïd mirror which allows the forward stimulating light to get through a hole and to focus backward stimulation light onto a photodiode. Another photodiode is located next to a sample mirror and the ratio of these two light measurements gives an indication about the insertion loss of the fiber link. The optical path of the stimulation light is depicted on fig. 4 while the OSL collection is depicted on fig. 5.

In a preliminary design, the housing was made from aluminum. New housings have been designed and patented to ensure tissue-equivalence of the sensor response regarding photon energy and angular incidence with respect to fibre axis [2]. A polymeric housing associated with tin cylindrical filters have been designed to make the dose measurement fit Hp (10) dose response in the gamma energy range [20 keV, 3 MeV]. Such a fiber sensor naturally provides circular symmetry. Only the angular incidence with respect to fiber axis is actually unconstant and requires compensation. With such a compensation concept, this angle may be as high as \pm 150 ° corresponding to a solid angle of 95% of space (*i.e.* 95 % * 4. π Sr), exceeding the standard of plane detectors.





4. Tests of OSL dosimeters

OSL sensors are calibrated with cesium and cobalt sources. A sealed cesium source is used (~ 1 mGy/h) for laboratory tests and a cobalt source is used for the final calibration of sensors (the dose rate ranges from 10 mGy/h to 100 mGy/h). The sensitivity depends both on the sensor and the acquisition unit. Therefore, there is as many calibration files as unit-sensor combinations.

The calibration of the first generation sensors (AES materials) involve several rounds of measurement corresponding to several orders of magnitude in dose rate (typ. ranging from 1 mGy/h up to 1 Gy/h). For each dose rate, OSL measurements are performed with varying integration periods to take into account fading effects in the calibration algorithm. Cesium and cobalt sources are pertinent calibration sources for dismantling operations because their photon energies (0,662 MeV and 1,25 MeV respectively) fit those encountered in these applications.

By contrast to AES, α -Al₂O₃ crystals are much less prone to fading at room temperature so that the calculation procedure of fading correction is not necessary.

The OSL response is obtained by integrating the OSL signal over time and subtracting the offset due to residual laser light level and Cerenkov scintillation. The offset subtraction method enables to separate the dose-rate effect (scintillation) from the dose effect (OSL). This feature is shown in fig. 6 from which one can see that the global OSL signal is shifted upwards because of the increase of scintillation light – consecutive to an increase in dose rate - incoming to the photo-multiplier.

The reset time of the material is inversely proportional to the intensity of the stimulation light. The reset time is about 2 minutes for an intensity of about 0,15 W/cm² (fig. 6). This reset time can be reduced by increasing the light intensity. This can be obtained either by increasing laser power or decreasing the exposed surface or both. By using alumina fibre crystals ($\emptyset = 1 \text{ mm}$, 10-mm long) instead of current crystal pellets, a 25-fold reduction in reset time is expected (*i.e.*, 5 seconds) with the same laser power.

The fig. 7 shows that the OSL response with respect to dose is linear (slope is unity in log scale) over four decades in dose. As can be seen with the log scale, the higher the dose, the lower the uncertainty of OSL measurement (%). The uncertainty in OSL measurement is due to the statistical error (proportional to $1/\sqrt{N}$ where N is the photon counts) and to residual light (laser light and scintillation). The error due to residual light is predominant for low dose measurements. The order of magnitude of the OSL dose resolution may be obtained by considering that the error is of same amplitude as the mean dose response. According to this definition, the fig. 7 teaches us that the resolution of the second-generation prototype equipped with a 20-meter long fibre is around 20 μ Gy.



FIG. 6 : Influence of dose and dose-rate on the optical signal

(the dose-rate is proportional to the offset (scintillation) while the dose (remaining OSL part) is proportional to the integral of the signal)



FIG. 7 : OSL response vs dose (OSL 2 system)

5. Applications

OSL dosimetry provides the user with a lot of advantages that are listed below :

- dose linearity and four orders of magnitude in dose range,
- remote dose and dose-rate measurements over long distances (up to some km for first-generation systems),
- electromagnetic immunity,
- tiny detectors (several mm³),
- intrinsic safety due to dose storage,
- quasi real-time operation (on-line stimulation),
- system handability and ease of use for non-specialists,
- chemical stability,
- low Z_{eff} for radiation protection by second-generation systems (compensation for photon energy and angular incidence),

Finally, the choice for a given OSL system will depend on the application and the required performances (resolution, temperature stability, ...), as well as whether a great fibre length or tissue-equivalence is required. The main applications are described in table 1 as ranked from high to low dose level (from left to right).

Sectors	Reactors	Storage sites	Industrial processes	Medecine	Nuclear installations	Radiation protection
Typical application	Generation of electricity	Underground storage of nuclear waste	X- or e-beam composite curing	Radiotherapy Radiology	Dismantling, decommissioning, exhaust monitoring	Personal dosimetry
Tissue- equivalence	NO	NO	NO	YES	NO	YES
Remote operation	YES	YES	YES	NO	YES	NO
Dose range	> kGy	>10 Gy	> 10 Gy	10 mGy – 10 Gy	0,1 to 100 mGy	µGy - mGy
Dose level	VERY HIGH	HIGH	HIGH	MEDIUM	LOW	VERY LOW

TAB. 1 : Applications for OSL remote dosimetry (classified according to high to low dose level)

5.1 Applications in process control

The OSL systems may find applications in the control of processes involving nuclear sources. For example, manufacturers of composite structures use electron beams or X beams for *in-situ* polymer curing (aeronautic applications). OSL sensors would provide these manufacturing companies with on-line remote distributed radiation sensing devices already calibrated for the given radiation source used.

OSL sensors may also be used for gammagraphy by inspecting the thickness and/or composition of unknown structures and in sterilization and food irradiation plants.

5.2 Applications in monitoring of underground or near-surface waste disposals

Nuclear waste conditioning is the last step in the nuclear fuel cycle. In France, three technical ways are undertaken to manage the nuclear long-life waste. The first way is transmutation in fast neutron reactor type (to turn long-life radioactive elements into short-life ones). The second way is near-surface storage and the third way is underground storage. In this latter storage condition, the waste canisters would be buried into disposal galleries interconnected *via* main galleries with a main access shaft to ground surface. A remote dosimetry may provide a useful remote dose monitoring before and long after installation in the near-surface storage conception and perhaps also in the conception of deep disposal sites. In the USA, a disposal site is under investigation in the Yucca mountains. In Germany, similar experiments are taking place in Gorleben salt mines. In France, an underground laboratory is under construction in Bure (Meuse).

5.3 Applications in radiation monitoring

OSL sensors may be fixed at many locations close or into nuclear reactors of power plants - either existing (PWR) or predicted (European Pressurized Water Reactor - EPR) - or of nuclear-powered submarines. In nuclear containments, optical fibres are particularly attractive to monitor radioactive pollutants in air flows because of their strong adaptability to existing filters.

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5.4 Applications in dismantling of nuclear installations

Several systems of the first generation have already been used by nuclear dismantling laboratories of the CEA (*Atelier Pilote de Marcoule*, CEA-Marcoule) and by COGEMA in order to make inspections of remaining activities after successive washes [7]. No commercial devices are actually able to deliver a real-time remote dose measurement through long small-diameter pipes. This OSL system was therefore of great help to make a dose cartography inside the pipe circuitry and tanks (fig. 8).

FIG. 8 : Use of the first generation OSL system for remote inspection of small pipes. (courtesy from CEA-DEN, SPRO CEA-Marcoule)



5.5 Applications in Radiation Protection

Workers in nuclear industry are very reluctant to being linked to an external acquisition unit. Telemetric systems are thus preferred in the perspective of real-time monitoring. Fiber remote radiation Protection of workers is therefore permanent rather than mobile. This radiation Protection equipment may be distributed on any structure of interest, for instance irradiation cells. Another application of interest is radioprotection of PetaWatt laser installations, for instance those of Livermore Laboratory in the USA or CEA-DAM in France. Very short-time laser bursts induce non-negligible X- and γ -irradiations inside the experimental chamber. Since the OSL technique provides a dose estimation, it is suited to the inspection of very short irradiations that would otherwise not be easily detected using electronic dosimeters. Moreover, an OSL equipment may be disposed off and remotely read without any maintenance procedure typical for TLDs.

5.6 Applications in Medecine (Radiology, Radiotherapy) : The MAESTRO European Project

At the beginning of the third millennium, one European citizen out of three will have to deal with a cancer episode in the course of his/her life. Worldwide, the estimated number of new cancer cases each year is expected to rise from 10 millions in 2000 to 15 millions by 2020. Within the European union, it is over 1,5 million new cancer cases that are diagnosed every year. Therefore, combating cancer is a major societal and economical issue for developed countries.

More than a half of all cancer patients are now treated by radiation therapy thanks to the technical progress made with irradiation equipment in the last ten years. For external radiotherapy for instance, high energy photon or electron beams are mainly produced by linear accelerators, while some limited number of proton synchrotrons are used for the treatments of cancers close to vulnerable organs (eyes, optical and auditory nerves, spinal cords).

Radiotherapy has now improved using the so-called Conformal Radiotherapy. In this concept, it is of prime importance to deliver the right amount of dose at the right place so as to kill the cancerous tumor while sparing most of the - still sound - surrounding tissues. High precision in target localization and in dose quantity are thus important challenges for the development of this technique.

In this context, an Integrated Project (IP) of the Sixth European Framework Program of the European community has been launched in 2004. The MAESTRO Project (*Methods and Advanced Equipments for the Simulation and Treatment in Radio Oncology*) is dedicated to the development of several technologies and treatment techniques in cancer Radiotherapy. Among these techniques, the OSL technique is now investigated in the purpose of providing doctors with quasi real-time, *in-situ* dose measurements along the patient body. The technical objective is to design an acquisition unit able to make a real-time dose cartography during the irradiation treatment. A conversational software will then give a real-time picture of the dose evolution to enable an accurate control of dose delivery.

Conclusion

Two generations of OSL systems have been developed at CEA during the past eight years. The first generation (OSL1) is mostly used for dismantling and process control whereas the second generation (OSL2) was developed for radiological purposes (radiation protection, radiology, radiotherapy).

The first generation relies on rare-earth-doped AES (MgS or BaS) and operates in the red/nearinfrared part of the spectrum. The second one relies on alumina crystals and operates in the blue/green part of the visible spectrum. These two systems exhibit a good linearity with more than four orders of magnitude range. The OSL1 system does not provide a tissue-equivalent dose measurement and is dedicated for dismantling operations and process control where a great fiber length is required (100 meters). Several systems of this kind are routinely used both by the COGEMA and the CEA for assistance in their dismantling operations. They provide remote, real-time, accurate, electromagneticimmune measurements and are suitable to operate in very hard-to-reach areas (*e.g.* pipes).

The OSL2 system has been developed for radiation protection purposes and may involve a patented compensation technique (both for angular incidence (95 % of space) and photon energies from 20 keV to 3 MeV). It exhibits a low fading and good sensitivity for low γ -dose measurements ($\approx 20 \,\mu$ Gy) with a typical 20-meter-long fibre length.

By now, only dismantling and decommissioning of nuclear installations are the most important applications for OSL systems. It is probable that in a near future OSL dosimetry will play an important role in medecine (Radiotherapy and Radiology) as well as in Radiation Protection of workers for which compensation methods have been designed and patented by the CEA.

Our further objectives – in the framework of the MAESTRO project - are to improve the portability of the instrumentation (size & weight), shorten the reset time by decreasing the size of the transducers and increasing laser power, optimize the cost of the optical acquisition unit, and incorporate an optical switch in the system to address several sensors in a parallel arrangement with a single acquisition unit. Since the sensor cost is negligible compared to the cost of the optical acquisition unit, the cost per measurement point may be considerably lowered using such configuration.

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