

## **In1 Situ, Long-term Monitoring System for Radioactive Contaminants**

James S. Durham (jim.durham@colostate.edu, (970) 491-0563)

Colorado State University

Department of Radiological Health Sciences

Fort Collins, CO 80523

Mark S. Akselrod (makselrod@ionet.net, (405) 377-5161)

Landauer Crystal Growth Division

723 ½ Eastgate

Stillwater, OK 74074

Steven W. S. McKeever (u1759aa@okstate.edu, (405) 744-5802)

Oklahoma State University

Department of Physics

Stillwater, OK 74074

### **Abstract**

The purpose of this project is to develop and demonstrate a low-cost, long-term *in situ* subsurface monitoring system for radioactive contaminant plumes. The monitor consists of a sensor based on optically-stimulated luminescence (OSL) readout of  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>:C (aluminum oxide) rods and a reader that interrogates the sensor at a prescribed frequency. Within a period of three years, this project will demonstrate, on a prototypical scale at a Department of Energy (DOE) site, a monitoring system that is capable of providing essentially real time measurements of the presence or absence of underground radioactive contaminant plumes.

The sensor will replace the need for repeated soil sampling around waste tanks, trenches, pits, cribs, and other sources or potential sources of gamma- or beta-emitting radioactive material. The sensor will also provide the Department of Energy (DOE) with a low-cost method of verifying, on a continuous basis, the absence of radioactive contaminant plumes and detecting changes in the activity of known contaminant plumes. The sensor will detect an increase in radioactivity at a given location or array of locations, thus providing a detection system for long term monitoring of waste tanks and trenches.

The sensor consists of a short (~ 1 cm) aluminum oxide rod that is physically and optically coupled to a conventional fiber optic channel. The sensor is placed in new or existing boreholes around radioactive waste sources such as waste tanks, trenches, and cribs. The fiber optic channels are sheathed in an inert material to prevent damage to the channel and to facilitate deployment in potentially hazardous environments. Aluminum oxide is an inert, structurally strong material that will not interact or be affected by hazardous materials that it may contact during deployment. A portable readout device, consisting of a light source, a photomultiplier tube, and associated electronics, will be used to measure radiation dose collected by the sensor. The measured dose is subsequently correlated to the type of radioactive contaminants and contamination levels in the soil by using appropriate calibration factors.

## Introduction

The Department of Energy (DOE) is the steward for hundreds of radioactive waste sites throughout the DOE complex. Most of these waste sites require remediation to stabilize the migration of radioactive constituents. A remote system that can monitor radioactive contaminant plumes would have application in numerous situations, including detecting and quantifying radioactive plumes during remediation efforts, verifying the absence of radioactive materials, and providing assurance that radioactive material is not leaking from remediated sites. The monitoring system described in this paper can provide DOE with a tool to monitor the contamination levels at active and remediated sites without requiring soil samples.

The goal of this work is to design, construct, and demonstrate a long-term, *in situ* monitoring system for subsurface radioactive contaminants. The monitor would provide data that would eliminate the need to obtain and analyze subsurface samples, resulting in a greatly reduced cost to DOE. The monitor consists of a sensor and a reader. The sensor is an aluminum oxide solid state dosimeter connected to a fiber optic channel and the reader consists of a strong light source, a photomultiplier tube (PMT), and a computer to record and store or transmit the measured data. The results of this work will provide a functional, prototype subsurface monitor, connectors for both a portable and stationary monitor, technical recommendations on the operation of the monitor, and calibration factors to relate the measured radiation dose to subsurface contaminant levels. This project will produce a low-cost monitor to measure radioactive plumes that is cost effective, provides near-real time measurements, continues to provide monitoring for many years, and that does not impact and is not impacted by the environment in which it is placed.

The subsurface monitor can be used to characterize and monitor waste tanks, trenches, and cribs at DOE sites during remediation planning and remediation activities through prompt detection of radioactive contaminants and after remediation is complete to verify the remediation process. Following remediation, the ability of the sensor to provide comprehensive automated monitoring reduces the cost of stewardship.

Operation of the monitor is as follows. The monitor is emplaced at a subsurface location and the radiation dose is allowed to accumulate for a prescribed period of time and the solid state dosimeter stores information that is proportional to the absorbed radiation dose. The length of time will vary depending on the expected or known radiation levels at the emplacement position. A reader will then be connected to the sensor. A light source in the reader sends a light signal through the fiber optic channel that causes the information stored in the dosimeter to be released in the form of a light signal with a wavelength that differs from the stimulation light. The emission light is converted to a voltage signal by the PMT, and the computer records the voltage signal. Through calibration, the emission light signal is related to the subsurface contaminant level.

This project will be completed in three phases. In phase one, the sensor will be produced and tested. Phase two will entail designing and testing the reader portion of the monitor, and phase three will focus on a field demonstration of the monitor at a DOE site to be identified as part of the project. The benefits of this project to DOE include a long term, *in situ* radioactive contaminant monitor that is easy to install, operate, and maintain. The use of the monitor will result in a decrease in the operating and maintenance costs associated with long-term monitoring

of radioactive contaminant sources and a reduction in spread of contaminants through prompt detection of radioactive contaminants.

## Technical Details

The subsurface monitor uses a process called optically stimulated luminescence (OSL) to read out a solid state dosimeter (SSD) using either a portable or a fixed reader. The sensor is a small crystal of anion deficient aluminum oxide which has the chemical formula  $\alpha\text{-Al}_2\text{O}_3\text{:C}$  in the form of rods with dimensions of 1 mm diameter and 1 cm long. The crystal is optically and physically attached to standard fiber-optic channel. The reader uses a laser or other strong light source, a PMT, and associated electronics including a computer to store and/or transmit the data to a remote location.

OSL is a physical process that has recently found commercial application in the field of personnel dosimetry. In order to understand the physics of OSL, a short discussion of solid state physics is necessary. In solid state physics, pure crystalline materials are modeled using band diagrams containing a valence band, a conduction band, and a forbidden band. The valence band is associated with electrons that form electronic bonds between atoms in the crystal. The conduction band is associated with electrons that are free to move about the crystal, enabling the crystal to conduct electricity. The forbidden band is associated with energy states in which electrons cannot exist in the crystal. If an impurity that has either one more or one less electron than the crystal matrix is introduced into the crystal, intermediate stable energy states within the forbidden band are formed. When radiation strikes the crystal, electrons are promoted from the valence band to the conduction band, and a small fraction of the promoted electrons are trapped in the intermediate energy states. The number of electrons trapped in the intermediate energy states is proportional to the amount of radiation energy absorbed by the crystal. If the crystal is heated, the electrons in the traps are again promoted into the conduction band, and these electrons return to the valence band by emitting a characteristic wavelength of light. The amount of light emitted during heating is proportional to the radiation dose absorbed by the crystal. When heat is used to depopulate the electron traps, the process is called thermoluminescent (TL) dosimetry.

Optical light can also be used to depopulate electron traps, and this process is called optically stimulated luminescence (OSL). In OSL, the crystal is stimulated using one wavelength of light and the crystal emits a different wavelength of light. In the case of aluminum oxide, the stimulation wavelength is yellow and the emission wavelength is blue, as shown in Figure 1. The advantages of OSL include the ability to stimulate the dosimeter remotely through a fiber optic channel, and the emission light can be collected through the same fiber optic channel. Filters can be used to separate the stimulation and emission wavelengths. One advantage of OSL over TL is that no heating is required and, therefore, no significant energy is imparted to environment. The OSL process is basis for Luxel® dosimeters that have been used by Landauer, Inc., for their personnel dosimetry service since 1999.

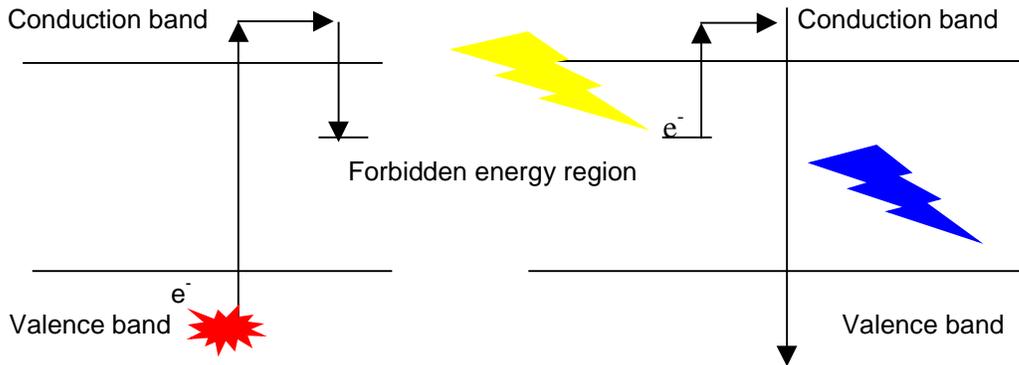


Figure 1. Energy band diagrams showing promotion of an electron by radiation into a trap (left figure) and subsequent stimulation and emission by the electron during readout (right figure).

Two different stimulation modes are possible. In continuous wave OSL (CW-OSL), the crystal is stimulated and the emission light is collected simultaneously (Figure 2). The two signals are separated using filters. A second readout method is pulsed OSL (POSL). In the POSL process, the crystal is stimulated for a short period of time (~100  $\mu$ sec) and the stimulation light is collected for the same short period of time in the absence of stimulation light. Shutters are used to shield the stimulation light and PMT so that the two signals are separated by time (Figure 3). Both readout techniques will be studied as part of this project. If the CW-OSL technique is used, the dosimeter will be annealed after each readout. If the POSL technique is used, the dosimeter will not be annealed after each readout and the dose history of the dosimeter will be stored.

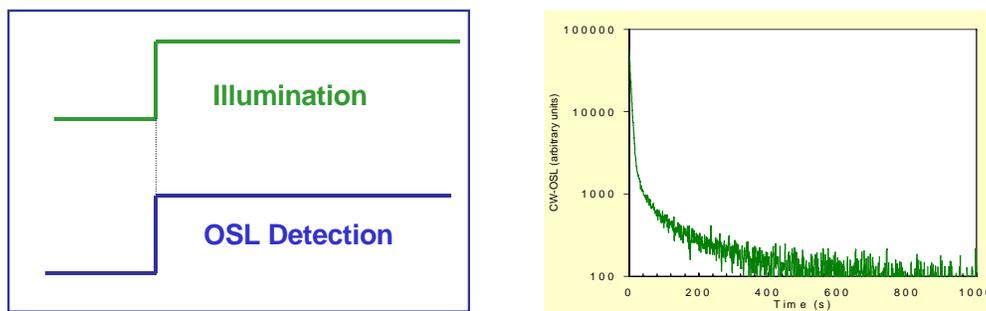


Figure 2. Schematic of stimulation for CW-OSL and the resulting output signal as a function of time. Note that annealing (erasing) of the signal requires nearly 100 s.

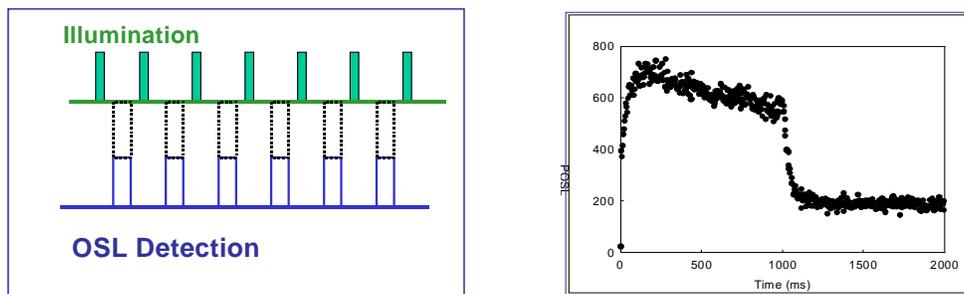


Figure 3. Schematic of stimulation for POSL and the resulting output signal as a function of time. The POSL process gives the same signal intensity as CW-OSL but is faster and does not anneal the dosimeter.

## Method

It is expected that the subsurface monitor will be used as follows. Multiple sensors will be placed in an array around a waste or remediation site at various depths using new or existing boreholes. The sensor will be allowed to accumulate dose for hours, days, weeks, or months depending on the contamination level of the subsurface soil. After the prescribed exposure time, a reader will be used to stimulate the dosimeter and the emission light will be collected. A calibration process will be used to relate the amount of emitted light to the soil contamination level. If a rise in the contamination level is observed, then the waste sight is leaking and further remediation is required.

Two readout options exist for the monitor. A technician can attach a portable reader to a single dosimeter and complete the measurement. Readout by this method is expected to require less than 5 seconds, and it is expected that this method will be used during remediation activities to locate areas of high contamination. A second option is to multiplex the sensors at a single, stationary reader. This would allow automated, continuous monitoring of a waste site by performing readout of many sensors at a single location. This method would be useful for long term monitoring of a remediated waste site.

The subsurface monitor can measure gamma radiation and moderate- to high-energy beta radiation. Using paired sensors will allow discrimination between beta radiation (emitted by  $^{90}\text{Sr}/^{90}\text{Y}$  or  $^{99}\text{Tc}$ ) and gamma radiation (emitted by  $^{137}\text{Cs}$ ). A cover placed over the sensor that is thick enough to stop beta particles but not thick enough to attenuate gamma radiation will allow the measurement of gamma radiation, while a sensor with no cover will measure both gamma and beta radiation. The gamma radiation contribution is obtained directly from the covered sensor, while the beta radiation contribution can be determined by subtracting the signal obtained from the covered sensor from the signal obtained from the bare sensor. Thus, the monitor can provide information on the type of contaminant.

## Technical Challenges

There are several technical issues that must be overcome before the subsurface monitor can be realized. The first challenge is attaching the dosimeter to the fiber optic channel. Methods to be studied to accomplish this task include the use of index-matched epoxy (to minimize light reflection at the interface) and fusion of the dosimeter to the fiber optic channel. The second challenge is to minimize light loss in dosimeter. A reflective coating will be applied to the exterior surface of dosimeter to allow collection of light that is directed away from the fiber optic channel.

The second technical challenge is to attach the reader to the fiber optic channel. A rugged, reusable connector will be developed that allows repeated connections between the reader and the fiber optic channel. In addition, a multiplexing connector will be procured and tested.

The final challenge is identifying an appropriate method of deploying the sensor. Several deployment strategies will be investigated, including the use of a cone penetrometer or existing boreholes.

## **Expected Results and Project Plan**

It is expected that the subsurface monitor will provide long-term monitoring of radioactive waste in the subsurface at a low-cost (\$50 target per sensor). The sensor will have a long life-cycle and will provide automatic, unattended monitoring with data archiving. In addition, a portable, hand-held reader will allow monitoring during remediation activities.

This project will result in the production of a functional, prototype subsurface monitor with connectors for both a portable and a multiplexed reader. In addition, this project will provide data from a field demonstration of the monitor and calibration factors to relate the monitor reading to the subsurface contaminant concentration. Technical specifications for the monitor will be provided including the cost of the sensor, the cost of the reader, the estimated lifetime of both the sensor and the reader, exposure time guidelines and deployment recommendations.

This project will be completed in three phases. During the first phase, the sensor will be developed. This will require growing aluminum oxide rods and fibers, attaching the dosimeter to the fiber optic channel, and demonstrating the proof-of-principle of the sensor through laboratory-scale experiments. These experiments will allow the estimation of the life cycle cost of the monitoring system and identify the sensitivity of the monitor. The measured sensitivities will be compared to sensitivities required for monitoring subsurface radioactive contamination plumes. A field demonstration site will also be chosen as part of the first phase.

The first phase will be considered successful when the sensor has demonstrated the ability to perform measurements with reproducible results for both gamma and beta radiation, obtain similar or relatable results for differing lengths of fiber optic channel, and can discriminate between beta and gamma radiation. Rod production will be considered successful when a minimum detectable dose of 1 mrem is realized.

The second phase of the project will focus on developing the reader. This will include demonstrating the proof-of-principle for both a portable and a multiplexing reader. Laboratory scale experiments will be performed to identify technical or economic issues with reader and rods. In addition, calibration factors will be measured that relate the measured signal to soil contamination levels for both beta and gamma radiation. Plans for the field demonstration of the sensor will be finalized, including completing any administrative requirements.

Success criteria for phase two include demonstrating the ability to show a relationship between contamination levels in soil and response of the sensor. In addition, a detachable connector that performs consistently after 100 removals and a multiplexing connector that measures dose accurately for five different sensors will establish success for this phase.

The final phase of this project will be dedicated to demonstrating the capabilities of the monitor at a DOE site. This phase will be considered successful when emplacement of the sensor is demonstrated, when repeated measurements of the same sensor provide results with no more than a 10% standard deviation, and when similar results are obtained for both the removable and multiplexed connectors.