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OPTIMIZED READOUT SYSTEM FOR
COOLED OPTICALLY STIMULATED LUMINESCENCE

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ABSTRACT

Cooled Optically Stimulated Luminescence (COSL) in $\text{CaF}_2\text{:Mn}$ is an ionizing radiation dosimetry method recently developed at the Pacific Northwest Laboratory (PNL) (1,2,3,4). In this method $\text{CaF}_2\text{:Mn}$ crystals irradiated by gamma radiation at room temperature are cooled to liquid nitrogen temperature (77K), stimulated by ultraviolet laser light at 326 nm, and allowed to warm to room temperature. Light emission proportional to the gamma exposure occurs as the TLD warms from liquid nitrogen temperature to room temperature. The new method is an example of a highly sensitive phototransfer technique which could form the basis for future radiation dosimetry applications. The COSL emission spectrum is similar to that reported by other authors (5,6). Measurements to date have shown high potential for measuring gamma exposures in the range of 10 μR . The high sensitivity of the COSL technique is due in part to the larger quantum efficiency of radiative recombination at low temperatures and to the complete absence of the incandescent background associated with conventional thermoluminescent readout methods. Along with the potential for a system which is more sensitive than thermoluminescent readers, multiple COSL readouts can be performed with minimal reduction in the COSL intensity. The multiple readout capability can serve as a possible permanent dosimetry record, thus allowing the reanalysis of a questionable reading.

In an attempt to optimize the sensitivity of the COSL method, a new readout system is being developed. The readout apparatus consists of a rotational stage which translates a $\text{CaF}_2\text{:Mn}$ dosimeter through a series of three stages. The function of the three stages are 1) loading, 2) cooling to 77K and ultraviolet laser stimulation, and 3) rapid heating through contact with a hot finger (in this case, a piece of metal at room temperature). A very low noise, high quantum efficiency phototube is used in the photon counting mode. The rapid heating of the sample is one of the major improvements of the new COSL readout system, serving to lower the phototube background interference. Photographs detailing the new readout system and a detailed explanation of the COSL process will be presented, along with recent data obtained with an older manual system.

OPTIMIZED READOUT SYSTEM FOR COOLED OPTICALLY STIMULATED LUMINESCENCE

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INTRODUCTION

For the past two years, the Pacific Northwest Laboratory (PNL) has been developing a new method of radiation dosimetry known as Cooled Optically Stimulated Luminescence (COSL) (1,2,3,4). The most sensitive material to date for COSL dosimetry has been thermoluminescent-grade $\text{CaF}_2\text{:Mn}$, commercially available from the Engelhart Corporation under the trade name TLD-400. The COSL readout technique of $\text{CaF}_2\text{:Mn}$ involves first irradiating the sample to gamma radiation at room temperature, next cooling the sample to liquid nitrogen temperature (77 K) in a liquid nitrogen bath, followed by illumination with an Helium-Cadmium (He-Cd) ultraviolet laser (326 nm) while still immersed in the liquid nitrogen bath. After this procedure, the sample is rapidly transferred to a readout chamber viewed by a photomultiplier (PM) tube. Photon counting of the sample is done as the sample warms to room temperature through thermal contact with the room temperature sample holder. Photon counting in multichannel scaling mode allows the registration of the luminescence as a function of time. The luminescent output as a function of time resembles the glow curve structures familiar in the more conventional field of thermoluminescent (TL) dosimetry. The integration of the glow curve is proportional to the gamma radiation exposure. Erasure or annealing of the

samples for reuse can be accomplished through either heating to 400 °C for 30 minutes, or by exposing the samples to an ultraviolet laser at room temperature (2). In this way, the COSL process provides a method for passive, radiation dosimetry similar to conventional TL dosimetry with a few differences.

COSL dosimetry provides the opportunity of sampling a fraction of the radiation damage centers, leaving the remainder of damage centers intact. The amount of radiation damage centers sampled depends on the amount of ultraviolet illumination received. The sensitivity of the dosimeter is controlled by the amount of laser excitation. Another obvious benefit of sampling a fraction of the radiation damage centers is the possibility of obtaining repeated measurements on an individual sample. The multiple readout capability can serve as a possible permanent dosimetry record with periodic inspection of the accumulated exposure without complete erasure of the exposure history. Another advantage of this physical property is the reanalysis of a questionable reading. As is well known, TL dosimetry involves the destruction of nearly all of the radiation damage centers during a single readout process.

The low temperatures associated with the COSL readout technique permit numerous advantages over TL dosimetry. The first obvious benefit of the reduced readout temperature is the elimination of the interfering incandescent radiation that accompanies a TL readout. (In the absence of an incandescent radiation background, a filter to eliminate this source of interfering light is not required.) This allows a closer coupling of the PM tube to the sample. Radiative recombination of electron trapping centers with their respective hole centers takes place with a higher quantum efficiency at low temperatures.

The probability for a recombination that produces a luminescent photon is therefore greater with COSL as compared with TL. For these reasons, the COSL technique offers the possibility for producing dosimetry systems that are much more sensitive than conventional TL systems.

In an attempt to optimize the COSL method, a new reader system is under development. The new readout system mechanizes an otherwise tedious readout process, while allowing the sample to rapidly warm to room temperature through contact with a hot finger. This paper will describe the development of the reader system, as well as the presentation of COSL data on $\text{CaF}_2\text{:Mn}$ taken manually with an older reader system (1).

EXPERIMENTAL RESULTS AND DISCUSSION

$\text{CaF}_2\text{:Mn}$ samples (0.636 cm x 0.636 cm x 0.089 cm) were annealed at 400 °C for 30 minutes. Following annealing, the samples were given exposures of 100., 500., 1000., 5000., and 10000. μR from a ^{137}Cs gamma source. After irradiation, the samples were stored in light-tight containers to prevent unintentional light exposure.

The samples were read out according to the COSL readout procedure outlined in Figure 1. Details of the manual readout apparatus can be found in a previous publication (1). The ultraviolet stimulation was provided by a 30 mW He-Cd laser emitting at 326 nm. The light exposures were performed while the samples were immersed in liquid nitrogen at a depth of approximately 1. cm. A fused-silica diverging lens expanded the laser beam to 0.7 cm in diameter, allowing the majority of the sample to be within the beam profile. A Uniblitz shutter enabled precise control of the laser energy that impinged on the sample. The actual readout data is displayed in Figure 2. Note the

exponentially decaying background, followed by a single luminescent peak structure. Figure 3 is a plot of the relative response of the samples as a function of 326 nm ultraviolet laser energy. Readout of the various exposures was done at the plateau region of the response curve in order to minimize response variation due to fluctuations in the laser energy. Response was determined by summing the photon counts in the peak.

Figure 4 is a log-linear plot of the COSL readouts for control, 100., 500., 1000., 5000., and 10000. μ R exposures. The laser was allowed to impinge upon the samples emersed in the liquid nitrogen bath for 5 seconds delivering a 150 mJ 326 nm exposure. Within the error bars of the measurements, the COSL response of the samples followed a linear relationship with ^{137}Cs gamma exposure. The COSL emission spectrum was measured and is identical to the TL emission spectra as measured by other authors (5,6). The emission spectrum is broad and peaks at 495 nm and can be found in Figure 5. Both the TL and the COSL emission spectra in $\text{CaF}_2\text{:Mn}$ have been attributed to a de-excitation of an electron from the first-excited $^4\text{T}_{1g}(^4\text{G})$ level to the ground state $^6\text{A}_{1g}(^6\text{S})$ level within the Mn ions (7,8).

Samples exposed to 50 mR were re-read without further gamma exposure or annealing to determine the multiple readout response. Figure 6 displays the results of these measurements. A 50% reduction was observed from the first to the second readout. Results obtained in earlier work (1) that utilized 254 nm stimulation, demonstrated approximately 10% reduction for the first few readings. Rereading of a dosimeter allows for the verification of a questionable result, while providing the possibility of a lifetime dosimeter. A single dosimeter could be issued to a radiation worker and analyzed periodically without an annealing step. The dosimeter would continue to accumulate exposure, while allowing periodic inspection. The COSL effect is

therefore well suited to perform the function required of a lifetime dosimeter.

OPTIMIZED READER DEVELOPMENT

The components of the optimized readout laptop system under development are displayed schematically in Figure 7. The system is controlled by a portable 386 personal computer. The computer houses boards for controlling the stepper motor and a multichannel scaling board used in recording the luminescent output of the samples as a function of time. Ultraviolet stimulation is provided by a 30 mW He-Cd laser. The laser is coupled to the sample via a fused silica optical fiber. The output of the laser into the fiber is controlled by a Melles Griot shutter that is activated by the computer. The samples are cooled through physical contact with the cold finger. The cold finger is supplied with liquid nitrogen by a 5 liter dewar. A solenoid valve is activated by the computer that allows delivery of liquid nitrogen from the pressurized dewar to the cold finger. A resistive thermal device measures the cold finger temperature. When the temperature rises above the set temperature, the solenoid valve is opened, allowing flow of liquid nitrogen to the cold finger.

A retractable drawer allows the sample to be inserted and removed from the reader. Once within the reader, the sample is moved from station to station by a plastic disk that is connected to the stepper motor. The first station is the cold finger, where the sample remains for a few seconds until the desired temperature is reached. After a few seconds the laser shutter opens, allowing the ultraviolet exposure to be delivered by the fiber optic. While the sample is on the cold finger, a constant light source is under the PMT. This feature allows for a calibration source to be counted with every

sample. After ultraviolet excitation is complete, the sample is rapidly moved to the hot finger. To subtend more light, the phototube sits above the hot finger, only 1. cm above the sample surface. The multichannel scaling board is activated as soon as the sample reaches the hot finger. After counting is complete, the system moves the sample to the first station. The sample drops into the bottom of the sample drawer, enabling the removal of the sample.

CONCLUSION

This data presented in this paper reveals the COSL technique is as sensitive as present TL techniques. The COSL technique offers the advantages of rereading of a sample, as well as the elimination of the incandescent background associated with heating. In an attempt to mechanize the COSL process, a versatile reader system is being developed to simplify and enhance the sensitivity of the process. The reader will allow for increased consistency of the COSL readout process, as well as faster warm-up time for the samples. Since the phototube background is linearly related to the counting time, reducing this variable will allow for more sensitive readings. When all the parameters of readout have been optimized for the new COSL reader system, it is conceivable a lower limit of detection of 10 μ R will be achieved.

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FIGURE CAPTIONS

Figure 1 - Flow chart outlining the COSL readout procedure.

Figure 2 - COSL readouts of $\text{CaF}_2\text{:Mn}$ exposed to control, 100., 1000., and 10000. μR ^{137}Cs gamma radiation. $\text{CaF}_2\text{:Mn}$ dosimeters were illuminated with 10 mW for 5 seconds. Readout was started 20 s after the 326 nm laser illumination at liquid nitrogen temperature.

Figure 3 - COSL response (output integrated for 8.0 s) as a function of 326 nm He-Cd ultraviolet laser excitation delivered to $\text{CaF}_2\text{:Mn}$ samples exposed to 50. mR from a ^{137}Cs gamma source. Each point represents the average of five readings minus control readings. Readout was started 20 s after 326 nm laser illumination at liquid nitrogen temperature.

Figure 4 - Net COSL response relative to ^{137}Cs gamma exposure in $\text{CaF}_2\text{:Mn}$ using 326 nm He-Cd laser illumination. The He-Cd laser delivered 30 mW of power to the samples for 5 seconds. The COSL outputs were integrated for 8.0 s. Net COSL response was determined by subtracting the average of five readings at each exposure level from the average of five control readings. Error bars represent 1 sigma.

Figure 5 - COSL emission spectrum of a $\text{CaF}_2\text{:Mn}$ sample. The emission spectrum is identical in the case of TL and COSL. Data was taken on a Tracer Northern spectrophotometer.

Figure 6 - Multiple COSL readout response as a fraction of the first reading. Each datum is the average of six measurements. $\text{CaF}_2\text{:Mn}$ dosimeters were

illuminated with 10 mw for 5 seconds. Readout was started 20 s after the 326 nm laser illumination at liquid nitrogen temperature.

Figure 7 - Diagram of the new optimized COSL reader under development.

CaF₂:Mn dosimeters were illuminated with 30. mW for 5 seconds. Readout was started 20 s after 326 nm laser illumination at liquid nitrogen temperature.

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