*COSL Dosimetry for Application to
On-Site Inspections:*

A Technical Review

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COSL DOSIMETRY FOR APPLICATION TO ON-SITE INSPECTIONS: A TECHNICAL REVIEW

by
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ABSTRACT

A technical review has been conducted of the cooled optically-stimulated luminescence (COSL) technique for high-dynamic-range dosimetry. Hardware and data presented by the technology developers have been examined. Applications of the COSL technology to tasks in treaty verification are identified, and possible problems in implementing the applications are discussed. Recommendations are presented for follow-on studies to resolve unanswered questions regarding the use of COSL in treaty verification.

EXECUTIVE SUMMARY

We have conducted a technical review of the cooled optically-stimulated luminescence (COSL) technology for high-dynamic-range dosimetry. Research on this technology has been supported by the Office of Arms Control and Nonproliferation (AN), on whose behalf the review was undertaken. The purpose of the review was threefold:

- (1) to assess the program conducted heretofore by the technology developers for its technical soundness and programmatic appropriateness;
- (2) to evaluate the potential for follow-on work on the technology, not necessarily to be funded by AN; and
- (3) to consider data taken with COSL relevant to warhead counting during re-entry vehicle on-site inspection (RVOSI), an arms-control-related scenario in which the technology is potentially applicable.

The review process involved a trip to Pacific Northwest Laboratories (PNL), the developers of COSL, for presentations and review of RVOSI data taken at the Los Alamos Simulation Facility (LASF). Subsequent to the review, COSL was used in warheadcounting demonstra-

tions at F. E. Warren Air Force Base, but data from those demonstrations were not available at the time of the review.

The following conclusions are drawn regarding the COSL work:

- The state of the work is accurately represented by progress reports and technical articles produced by the PNL group.
- The COSL technology, in its present form, remains essentially a laboratory-based system more suitable for R&D than for real-world applications. However, it appears likely that attempts to package COSL for real-world use, probably in conjunction with a commercial partner, would lead to success.
- In the LASF work, COSL did observe inhomogeneities in the gamma-radiation field related directly to the presence of three warhead mockups. However, the nature of those inhomogeneities was at odds with the results of other warhead-counting demonstrations against the same mockups or similar realistic objects. The source of these discrepancies apparently lies in the uncollimated nature of the detectors used in COSL, as opposed to the collimated detectors used in the other

studies. However, a detailed understanding of the signature obtained with COSL is not in hand, and real-world use of COSL for RVOSI will probably require additional basic research on the nature of the signatures, as well as the obvious implementation effort.

I. INTRODUCTION

As part of its funding strategy, the Department of Energy's Office of Arms Control and Nonproliferation (AN) calls for technical reviews of certain key technologies, whose development it has supported, to determine their state of development and future prospects. Such a technology is the cooled optically-stimulated luminescence (COSL) approach to dosimetry under development at the Pacific Northwest Laboratories (PNL). COSL is interesting as a possible technical asset for supporting arms control and nonproliferation because of its potential to improve on existing dosimetric techniques (for example, thermoluminescent dosimeters, or TLDs) in various ways, for example, dynamic range, data preservation after readout, etc. These potential improvements suggest a possible role for COSL in monitoring scenarios in which radiation detection is a key component.

The COSL technology has been described in the technical literature, and a comprehensive review of the physics principles involved will not be attempted here. The basic underlying principle of operation is that defects generated in the dosimeter material by radiation exposure can be caused to luminesce upon excitation with ultraviolet or blue light, the intensity of the luminescence being proportional to the number of defects present, which, in turn, is proportional to the amount of radiation received. This principle stands in contrast to the usual TLD approach of activating the material by heating it. Because the defects are not annealed out when excited by the laser, the COSL material retains a "memory" of the dose received, in contrast to TLDs, which are annealed during the readout process and, thus, lose their memory of the radiation exposure.

A specific application of the COSL technique has been proposed in the form of emplacement of an array of COSL dosimeters about an ICBM to determine the number of warheads it carries. This application is motivated by the possibility that the visual approach to re-entry-vehicle on-site inspection (RVOSI), as prescribed in the START treaty, might eventually be augmented or supplanted by instrumental approaches. Experiments performed at the

Los Alamos Simulation Facility (LASF), operated under AN funding at Los Alamos National Laboratory (LANL), and at F. E. Warren Air Force Base in Wyoming, have addressed COSL's feasibility for this purpose; certain of these experimental results will be discussed in the following text. Other applications have been discussed involving emplacement of COSL dosimeters "as a fire and-forget" means of monitoring movement of radioactive materials, but specific AN-driven experiments comparable to the LANL and Warren experiments have not been performed.

During FY 92, AN asked personnel from LANL and Idaho National Engineering Laboratory (INEL) to perform the COSL technical review. The authors traveled to PNL for this purpose in July 1992 to observe the hardware and resolve issues regarding its use in the RVOSI scenario. This report documents their findings.

II. COSL TECHNOLOGY

The COSL technology has two general components: the dosimeters themselves and the laser-based, cryogenic system for readout. The dosimeters consist of calcium fluoride (CaF_2) crystals doped with a small amount of manganese. Depending on the application, they are attached to various materials for emplacement; in the case of the RVOSI experiments, they were placed on metal hoops that fit around the item being inspected. In any case, these materials are application-specific and not tied to the technology itself.

The readout system consists of a PNL-designed cryogen for cooling the dosimeters to the liquid-nitrogen temperatures required for readout; a laser for exciting the luminescence; electronics for counting the luminescence photons; and a computer for recording the photon counts and controlling the movement of dosimeters into and out of the cryogen. In the current, benchtop model of the COSL apparatus, the electronics reside (apart from the photon counters themselves, which are within the cryogenic unit) in a standard NIM bin; presumably, task-specific electronics could be fabricated to replace this in the event that COSL should be commercialized. The system currently requires an external liquid-nitrogen supply for the cryogen, in turn, requiring a Dewar flask or other cryogenic container. In a future, commercialized system, this could possibly be replaced by a Stirling-cycle refrigerator, but it appears outside the scope of the current work to incorporate this technology.

In addition to the detection/data-reduction technology, a third component is required if the dosimeters are to be re-used: a heat source sufficient to anneal the dosimeters and thus remove the luminescent response. At the present time this function is served by a standard laboratory oven. This component is not part of the COSL technology *per se* and might or might not be required as part of the eventual COSL package, depending on whether the dosimeters were to be re-used. However, the dynamic range of the dosimeters is so great that background radiation might be readily detectable after several hours of exposure, and consequently, for the most sensitive applications of the technology, the detectors will probably have to be re-annealed just before use.

Some operational characteristics of the current COSL system are

- wavelength of photons produced by the interrogating laser: 326 nm;
- time to interrogate and read out each dosimeter: less than 1 min;
- liquid-nitrogen use rate: ~ 1 L/h; and
- time and temperature required for annealing: 400 °C for 30 min.

COSL has also been demonstrated with slightly longer-wavelength (351 nm and 406 nm) lasers, which offer certain operational tradeoffs that will be discussed later. Liquid-nitrogen usage is relatively high and might be improved in an eventual, field-portable system, even if the liquid-nitrogen system is not replaced outright by a Stirling-cycle refrigerator.

We discussed the studies performed on this system with the PNL technology developers, whose visual aids comprise the Appendix to this report. Some of the key findings are the following:

- Experiments have been performed to determine the dynamic range of the COSL technology — that is, the range of radiation exposures over which the dosimeter response is proportional to the dose received. Based on these experiments, the technology appears capable of correctly registering doses as low as 40 μ rad and as high as several times 10^4 rad. Claims of correct response down to the 10- μ rad level are neither supported nor contradicted by the existing data; in any event,

measurements below the 100- μ rad level are unlikely, in our judgment, to be of much use in verification scenarios because of the difficulty in making them in a background-free environment.

- The developers have investigated the uniformity of response of the CaF_2 crystals; that is, the degree to which different crystals respond differently to identical irradiations. The claim that the responses vary by $\pm 10\%$ from crystal to crystal appears justified by most of the data, but the experiments at the LASF may provide evidence for greater variations, as will be discussed below.
- Stability of the dosimeter response — that is, the degree to which the response will be independent of how much time has elapsed between irradiation and readout — has also been studied. For a 10-rad initial dose, it was found that there was a 4% fading in the response when readout was delayed for a year, compared to readout on a similar dosimeter done immediately after the irradiation. This compares favorably with the $\sim 10\%$ /month fading occurring with usual TLDs. We did not see any data addressing stability of response to smaller irradiations, which may be of more practical interest in verification scenarios.
- The developers have investigated the sensitivity of dosimeter response to energy of the irradiating photons. The detection efficiency for photons with energies below 100 keV is significantly greater than for higher energy photons, which is to be expected given the small size of the dosimeters. This may cause problems in interpreting the results of measurements in which the energy spectrum of the irradiating photons is not at least broadly known. Of course, COSL has no intrinsic spectroscopic capabilities, so that some other technology — or *a priori* calculations (guesses?) — would be required for gaining this knowledge. The significance of this observation in verification scenarios is unclear; under cooperative conditions no problems are likely to result, but spoofing scenarios involving low-energy gamma-ray sources might possibly appear in some cases. Investigation of these spoofs exceeds the bounds of this report, however. We noted during the review that the ratio {low-energy response}/{high-energy response} might be directional in nature, since the dosimeters obviously are not spherical. Experiments and Monte Carlo calculations to study the directionality of the dosimeters might be useful at some point.

- The claimed capability to read out COSL dosimeters several times (in contrast to conventional TLDs, which are annealed in the readout process so that the response information is lost) has been investigated experimentally, using a laser operating at 351 nm, rather than the 326-nm laser used in the standard system. The dosimeters still respond to laser illumination even after they have been read out as many as 6 times previously, although the response on the seventh readout is down to 20% of the initial response (that is, only 20% as many luminescence photons are produced per rad of dose received as the first time the same dosimeter was read out). Assuming a uniform decay in response with each readout, the loss of response per readout is then <25%. Statistics regarding the uniformity of this loss were not presented. If the loss is uniform from crystal to crystal, then as long as the readout history of a given crystal is known, the results should continue to be reliable for initial exposures of 1 mrad or greater even when the dosimeter is read out 7 times. If the loss is not uniform, reliability might start to be compromised after two or three readings.
- The developers have investigated the dependence of the readout upon the wavelength of the interrogating laser light, by interrogating previously-irradiated dosimeter materials with light from several different lasers (the normal 326-nm laser, and others producing photons of wavelengths 351 and 406 nm). Luminescent response decreases with increasing wavelength of interrogating photons, but this falloff is compensated by decreased "dark current" — that is, light escaping the dosimeters that does not result from irradiation-related luminescence. The optimum wavelength for the interrogating laser is therefore not totally clear and may be application-dependent.

The general conclusion of these observations is that COSL appears viable as a dosimetric technique and that the technology may be appropriately characterized as proven for this purpose. A few detailed questions still exist regarding the performance of the technology, but it is not obvious that these questions are critical considering the general state of COSL maturity. Given this generally satisfactory technical status, we turned to evaluation of the application of COSL in the RVOSI scenario.

III. RVOSI OPERATIONAL CONCEPT

The operational concept for the RVOSI application has the following general shape:

- A belt bearing recently annealed dosimeters is emplaced around the monitored object (missile, shroud, etc.) and left in place for some minutes to hours.
- After exposure to the monitored object, the belt of dosimeters is processed through the reader system, and the readings are saved in a computer.
- The results of the ensemble of dosimeter readings are interpreted to determine whether warheads beyond the attributed number are present. (The details of this interpretation have not yet been worked out and are undoubtedly scenario-dependent; the process may involve computer analysis for pattern matching, but possibly could also be done with simple visual inspection.)

This concept resembles the approach taken by the RAPID, PGS, and PNS techniques used in the warhead-counting demonstrations at F. E. Warren Air Force Base in late 1990. The key difference between the concepts is that, whereas the techniques used at Warren involved a single detector system* that was moved around the perimeter of the missile, taking one relatively brief reading at a time, the COSL approach uses multiple detectors at a time. Another difference was that the gamma-ray approaches used at Warren (PGS and part of RAPID) used collimated detectors, while the COSL detectors are intrinsically uncollimated, although collimation might be added at the expense of some operational complexity. The COSL detectors used in the work described below were uncollimated, in any event.

A test of this concept was performed using resources at the LASF, and results were presented at the site visit during the technical review. For that test only half of the measurement positions were used, covering a 180-deg arc on one side of the usual array of SS-20 mockups (Fig. 1). With the benefit of hindsight, a full 360-deg coverage would have been more useful, as it would have answered some of the questions associated with data interpretation

* Except in the case of RAPID, which used a single gamma-ray detector and two neutron detectors, the two neutron detectors may be considered to have operated as a single unit for the present discussion, as they were nearly collocated.

Fig. 1. Schematic diagram of experimental arrangement at LASF. The dark squares represent assemblies containing three COSL dosimeters each.

of a well-rehearsed demonstration at Warren, this viewpoint is understandable. We gather that, at Warren, the plans were for the belt of dosimeters to span the full 360 deg around the missile, but that an unexpected operational constraint reduced the coverage to a 320-deg arc. Again, the Warren demonstration did not occur until after the site visit, so data from that demonstration are not included in this report.

IV. REVIEW OF LASF RESULTS

Results of the LASF experiments were presented at the site visit and are shown in Figs. 2 and 3. The plots represent radiation received by the dosimeters as a function of angle around the ensemble of SS-20 mockups. Superimposed on the data are circles representing the locations of the radiation emitters in the mockups. Only two radiation emitters are shown, as the third mockup was positioned away from the arc of dosimeters as shown in Fig. 1.

These plots present some puzzling features. Peaks and valleys in the doses are seen, as would be expected for an inhomogeneous radiation field. However, the highest dose rates apparently are observed at dosimeter positions "between" the mockups; that is, at positions marked with

arrows in Fig. 1. This observation is completely at odds with the findings of RAPID and PGS at Warren, where the highest count rates (equivalent to dose rates) were observed in the detectors nearest the weapons inside the Peacekeeper missile. Although neither RAPID nor PGS were used against the SS-20 mockups in geometries similar to those in the COSL experiment, it is completely reasonable to expect that those techniques would have observed higher count rates at locations close to the SS-20 mockups as well.

A possible explanation for this discrepancy, proposed by the technology developers, is the uncollimated nature of the COSL dosimeters. Because COSL records radiation striking the dosimeters from all directions, the technique is sensitive to *all* gamma rays leaving the assembly of warheads/mockups. RAPID and PGS, by contrast, use collimated detectors and, therefore, detect primarily gamma rays traveling along the axis of the collimator, which normally is pointed at the center of the assembly; the detectors, in essence, have a "field of view" located along the axis of the collimator whose size is a function of the degree of collimation. Only sources within this field of view contribute gamma rays that are detected, assuming perfect collimation. With uncollimated detectors such as the COSL dosimeters, by contrast, the "field of view" is essentially 4π , and radiation is received and recorded from all sources present; variations in observed dose result from $1/r^2$ effects and the shielding of one warhead/mockup by another. These variations need not, and in general do not, correspond exactly to the variations arising from the field-of-view effect.

This explanation, while intuitively appealing, is unsatisfying in its implications for the usefulness of COSL (or other uncollimated dosimetric techniques) in RVOSI, because two problems of data interpretation immediately present themselves. First, unless the size of the radiation-emitting parts of the warheads is known, it may be difficult or impossible to determine *a priori* whether a peak or valley should be present near a warhead, as it may be unclear if the warheads shield each other at a given dosimeter location. Second, the COSL dosimeters are at least slightly directional (because they are not spherical), complicating the *a priori* prediction further. There being a presumed benefit to *knowing* how the technique will work in a given scenario, these uncertainties appear undesirable.

A second puzzling point in the LASF experiments concerns the scatter in the data. The COSL implementation used at LASF included three individual dosimeter chips at each location. The plot shown in Fig. 2 displays the

Fig. 2. Plot of radiation received by dosimeters in the LASF experiment. Points represent the average of the values exhibited by the three dosimeters at each angle. The open circles represent the locations of the two SS-20 mockups nearest the arc of dosimeters, as shown in Fig. 1. (Figure courtesy of S. Miller.)

Fig. 3. As in Fig. 2, but values for each of the three dosimeters at each angle are shown. (Figure courtesy of S. Miller.)

average of the doses received by the three different chips at each location, so that only one data point is presented at each angle. While the detailed meaning of this information is unclear, for reasons discussed in the preceding paragraphs, it does seem reasonably clear that some regular structure exists in the data. However, dose rates recorded by individual chips seem to show considerable scatter (Fig. 3). It is difficult to decide whether the scatter results simply from counting statistics or is rooted in some fundamental, inadequately understood property of the detectors (for example, poorer uniformity in response than expected), not least because it is unclear what the concept of statistical error means for dosimetry. However, for at least some of the locations, the disagreement among the three chips strikes one on first examination as inexplicably large. The technology developers have discussed the scatter with statisticians who assert that the scatter is not really unexpected; but lacking either the time to meet with those statisticians or the expertise to evaluate the statistical arguments, we were unable to assess their claims. The scatter seems worthy of additional investigation in any case.

V. CONCLUSIONS

We draw the following conclusions regarding the status of COSL at this time:

- **The state of the work is accurately represented by progress reports and technical articles produced by the PNL group.**

The technology developers have conducted a series of experiments demonstrating that the COSL technology “works”; that is, the signal produced at readout corresponds to the radiation dose received. The experiments appear to have been performed and interpreted correctly. Progress in bringing COSL to mature status has been reported in AN quarterly reports and in professional publications and appears to reflect the equipment, data, and procedures that we saw.

- **The COSL technology, in its present form, remains essentially a laboratory-based system more suitable for R&D than for real-world applications. However, it appears likely that attempts to package COSL for real-world use, probably in conjunction with a commercial partner, would lead to success.**

The use of cooled CaF_2 chips and a laser-based readout system appears reasonably mature. However, the

existing system consumes liquid nitrogen, a major drawback for many real-world applications, and uses a laser for readout that may not be optimized for such applications. There exists cryogenic apparatus that could probably substitute for liquid-nitrogen cooling in a final, fieldable system; incorporating such apparatus lies beyond the scope of the work proposed and undertaken by the PNL team, but is a reasonable component of a commercialization of COSL. The laser used in the current system is not particularly large or complex, as lasers go, but it is still relatively large for field use and requires ac power that may not be available in the field. A battery-powered laser or bright, LED-based blue light source may or may not be feasible as a system component, but in any event is the province of a commercial partner rather than the technology developers.

- **In the LASF work, COSL did observe inhomogeneities in the gamma-radiation field related directly to the presence of three warhead mockups. However, the nature of those inhomogeneities was at odds with the results of other warhead-counting demonstrations against the same mockups or similar realistic objects. The source of these discrepancies apparently lies in the uncollimated nature of the detectors used in COSL, as opposed to the collimated detectors used in the other studies. However, a detailed understanding of the signature obtained with COSL is not in hand and real-world use of COSL for RVOSI will probably require additional basic research on the nature of the signatures, as well as the obvious implementation effort.**

This conclusion appears self-explanatory.

We recommend the following actions as appropriate follow-ons to the COSL work, although it is not clear that they are appropriate for AN funding:

- Some additional effort to characterize the chip response might still be worthwhile, as a hedge against applications (RVOSI or others) where either the gamma-ray energy spectra and/or the source locations/sizes are unknown. Examples are studies to understand the directionality of the chips and the variation in response with gamma-ray energy, possibly involving production and evaluation of chips with different dimensions than those used in the current system. These studies would be particularly valuable when combined with the Monte Carlo modeling that we also recommend below.
- Explorations of blue- to ultraviolet-light sources other than the existing lasers for stimulating the luminescence could provide a basis for commercialization of a more fieldable COSL system. Some of this work has already been done, according to the technology developers, but more work appears warranted.
- It appears important to understand the LASF experiments more fully, not so much because RVOSI is a particularly critical application of COSL (it is not) but because the LASF work is a prototype of arms-control-oriented activities in which COSL could possibly contribute — but only if the possible contribution is well understood. Consequently, Monte Carlo modeling of the LASF work, including parallel modeling of the work with collimated detectors if applicable, would appear to have some value.

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