# FINAL REPORT

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# *MONOLITHIC INTEGRATED RADIATION SENSOR USING STIMULATED LUMINESCENCE FROM ALUMINA*

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## SUMMARY

The project goal was to design and test a monolithic integrated device for radiation sensing, using optically stimulated luminescence (OSL) from $Al_2O_3$:C. The device would consist of GaN/InGaN-based components epitaxially grown on each side of a $Al_2O_3$:C substrate. Radiation energy stored in the substrate would be stimulated by visible emission from a GaN light-emitting diode (LED) grown on one side of the device, and the OSL emission from the substrate (in the blue region of the spectrum) would be detected by the InGaN p-i-n diode grown on the other side of the substrate. The primary application of the device would be in space radiation environments. Thus, two major research thrusts were launched during this project. Firstly, research at Oklahoma State University (Dr. Stephen W.S. McKeever and Dr. E.G. Yukihara) concentrated on characterization of the OSL properties of $Al_2O_3$:C in radiation fields typical of those experienced in low-Earth orbit. Secondly, research at the University of Washington (Co-Is, Dr. T.G. Stoebe and Dr. T. Chen) focused of device development and GaN/InGaN epitaxial growth. While progress in each line of research has been substantial, the ultimate goal (that of producing a working prototype device) has not yet been reached. In the following sections we detail the research progress and identify outstanding issues.

## 1. RADIATION CHARACTERIZATION OF OSL FROM $AL_2O_3$:C (Oklahoma State University)

### *1.1 Objective:*

The objective of this part of the project was to characterize the OSL response of aluminum oxide dosimeters to high-energy heavy charged particles (HCPs) for space dosimetry applications. We intended not only to achieve a better understanding of the physical mechanisms responsible for the OSL to low and high Linear Energy Transfer (LET) particles in this material, but also to develop a method that allows the discrimination between the dose due to the low and high-LET part of the spectrum of high-energy charged particles in space.

### *1.2 Primary Irradiation Facilities:*

Single and mixed field HCP irradiations were carried out primarily at the Heavy Ion Medical Accelerator (HIMAC) at the National Institute of Radiological Sciences (NIRS) in Chiba, Japan, either as stand-alone experiments or as part of an international intercomparison program known as ICCHIBAN. Additional exposures were carried out at the NASA Space Radiation Laboratory (NSRL-ICCHIBAN) at Brookhaven, at the Loma Linda medical accelerator (Proton-ICCHIBAN), and at the Autonoma University of Mexico (UNAM) Pelletron in Mexico City (for low energy protons and He ions). Additionally, several space-based exposures were carried out during several Shuttle, Soyuz and ISS flights. Specific experiments included flight STS-105 (July '01), ISS (July-Dec. '01), BRADOS (ISS, Oct.-Dec. '03) and MESSAGE (ISS, Oct.-Dec. '03). Finally, high altitude balloon flight exposures were also performed as part of the TRACER (Dec. '03) experiment. Samples were also prepared for the MATROSHKA experiment (currently on the ISS).

### *1.3 Measurements and Findings:*

The OSL of dosimetry grade $Al_2O_3$ is already established for $\gamma$, beta and x-ray dosimetry. Different forms of $Al_2O_3$ crystals (and, for comparison, LiF:Mg,Ti and $CaF_2$:Tm) were exposed to several HCP beams ($^4$He 150 MeV/u, $^{12}$C

400 MeV/u, $^{12}$C 135 MeV/u $^{20}$Ne 400 MeV/u, $^{28}$Si 490 MeV/u, and $^{56}$Fe 500 MeV/u, protons of various energies and mixed fields, including beta ($^{90}$Sr) and gamma ($^{137}$Cs) irradiation). The HCP energies corresponded to LET values ranging from <1 to 189 keV/μm (in water). The results indicated that the OSL response depends upon a number of material and experimental factors and on the energy and type of particle used in the irradiation. Different OSL measurement modes – namely, continuous wave OSL (CW-OSL) integrated area, CW-OSL peak intensity, and pulsed OSL (POSL) - result in different OSL efficiencies for the same particles, and in different OSL dose response curves. In general, the relative OSL sensitivity (compared to that from $^{60}$Co gamma irradiation) decreases as the LET of the charged particle increases. Thus, it is not possible to accurately determine the absorbed dose D in luminescence detectors irradiated in unknown particle fields (or the dose equivalent, H) if the variations in the efficiency are not considered. The previously observed ionization density dependence of the OSL curve of $Al_2O_3$ was confirmed in our experiments and this opened the possibility of developing methods to extract information about the radiation quality or to discriminate between low and high-LET radiation in a mixed fields such as those found in space. One possibility we have examined is to extract parameters from the OSL curves from each radiation type in order to estimate a "mean LET". This information, in conjunction with the "mean efficiency" allows an estimate of the absorbed dose D. Less accurate, however, is the determination of H by this method.

An alternative approach that we are currently in the process of developing is a deconvolution method in which the OSL signal from the low LET part of the radiation field can be separated from that due to the high LET part of the field. Preliminary tests at OSU using "simulated" OSL signals from mixed fields (i.e. a linear superposition of OSL curves due to individual particles in known proportions) has resulted in encouraging results. In this way the low-LET dose may be evaluated using OSL while the high-LET dose may be evaluated using other means (e.g. plastic nuclear track detectors, PNTDs). All of the above promising results require further investigation and development.

### *1.4* ***Future plans:***

The second year of the present research project includes experiments to improve our understanding of the OSL efficiency *versus* LET curve of $Al_2O_3$ and to test the deconvolution procedures elaborated above, primarily based on irradiations at HIMAC. Some of the objectives are to determine whether or not the response of $Al_2O_3$:C can be represent by a single curve as a function of the LET, and to test the deconvolution techniques for separating the components with lower and higher than 10 keV/μm.

## 2. MONOLITHIC DEVICE DEVELOPMENT BASED ON GaN/InGaN EPITAXY ON $Al_2O_3$ SUBSTRATES (University of Washington)

### *2.1* ***Objective:***

The objective was to design and develop a miniaturized radiation dosimetry system based on an earlier integrated sensor concept to integrate a radiation sensor based on optically stimulated luminescence (OSL) of $Al_2O_3$:C by using the $Al_2O_3$:C as the MOCVD growing substrate for a high quantum-efficiency InGaN-based p-i-n photodiode. This provides the base for a dosimeter with a built in luminescence detector. Add a light-emitting diode (LED) on the opposite side as the stimulation source, and one would have a fully integrated radiation sensor, as illustrated in Figure 1.

An issue with all current OSLDs (and TLDs), however, is that current systems require that the OSL dosimeter be dismounted from its badge for reading in a separate "reader." The device proposed in this proposal, however, has the reader built-in, will consume very little power as compared to current systems, and will be of extremely low mass (a few grams). The intimate contact between the excitation source and the dosimeter on the one hand, and the detector and the dosimeter on the other, will ensure high stimulation and optical detection efficiency for the OSL.

**Figure 1. The illustration of the proposed radiation sensor**

## *2.2 Simulations:*

### *The Responsivity Performance of GaN Photodiodes*

The responsivity performance of the novel GaN devices have been simulated by ATHENA and ATLAS, which were developed by SILVACO International, Inc. ATHENA was employed to define the structure of GaN p-i-n photodiode. All the parameters of GaN were obtained from published articles., ATLAS was used to evaluate the characteristics of the photodiodes both with and without illumination.

The optimal structure of GaN p-i-n photodiode was investigated. With zero bias voltage, the depletion width can be determined to be 0.65 micron. Thus, the width of the i-region, $W_i = 0.6$ micron was used to ensure that the i-region is completely depleted. The effect of $W_p$ on the responsivity of the GaN photodiodes was studied. From simulation, the responsivity is defined by the ratio of cathode current to optical power. $W_p$ was varied from 0.1 to 0.7 micron, with other parameters held constant. The highest responsivity at 360 nm of 0.27 A/W was achieved with $W_p$ =0.25 □m. Due to the sharp band edge and the high absorption coefficient (□$_□$ = $10^5$ cm$^{-1}$), only $W_p$ has a significant effect on the spectral responsivity. The simulated result of the peak responsivity is 0.23 A/W at 365 nm on $W_p = 0.2$ □m.

The simulated result of the detection response of the GaN p-i-n photodiode with $W_p = 0.25$ □m showed that the detectable wavelengths of the GaN p-i-n photodiode avoid detection at 470nm from LEDs.

The responsitivity of the GaN p-i-n photodiode was also simulated the following parameters:

p-region; hole concentration = $8\times10^{18}$ cm$^{-3}$; thickness $W_p = 0.25$ □m
i-region; electron concentration = $4\times10^{16}$ cm$^{-3}$; thickness $W_i = 0.25$ □m
n-region; electron concentration = $5\times10^{18}$ cm$^{-3}$; thickness $W_n = 1.05$ □m

The responsivity of the GaN p-i-n photodiode indicated that there were two peaks contributed by the UV and F-center emissions of the OSL from $Al_2O_3$:C. After de-convoluting both peaks, the responsivity from each emission can be resolved, in this case, sixty six percent of the responsitivity of the GaN photodiode was from UV component of the $Al_2O_3$ OSL emission and 34% of the responsitivity of the GaN photodiode was from F-center emission of the $Al_2O_3$.

By comparison with the performance of commercially available OSL systems, the minimum detectable dose (MDD) of the new sensor was estimated to be approximately 0.06 mGy

### *Simulation of responsitivity of GaN photodiodes under radiation:*

For the real-time applications of the new radiation sensors, it is essential to study the noise level caused by the radiation on GaN photodiodes. A calculation has been conducted to obtain the noise level of x-ray (energy at 12.4 keV) and gamma ray (energy at 1 MeV) on GaN photodiodes by University of Washington (UW).[17] The results indicate that the responsivity of GaN photodiodes was $8.04\times10^{-7}$ A/W at an energy of 1 MeV and $6.49\times10^{-5}$ A/W at an energy of 12.4 keV with dose rate of 0.1cGy/s as shown in Table 2. The calculations indicate that the radiation rate of 0.1cGy/s of gamma and x-ray would not generate significant noise during device operation.

## *2.3 Prototype Testing Results:*

Preliminary experiments with the new sensor have been undertaken on a prototype by using a Si photodiode (Hamamastu S1787) packaged with a proper filter (narrow band 365nm), which has 5 to 10 times lower responsibility than GaN detector in the UV range, as the detector device; an $Al_2O3$ OSL dosimeter; and a LED (Nichia NSPG300A) as a stimulation source. Each component was in direct contact with the neighboring elements of the system. A computer with data acquisition software collected the output signal from the detector. The result demonstrates the feasibility of the new dosimeter system, with the system able to detect to the dose at the 100 mGy level and better. With improvement to and optimization of the components, such as wavelength matching between OSL dosimetry material and the GaN photodiodes, and a more sophisticated circuit for low level current capture, the sensitivity of the sensor is expected to be at the range of mGy and lower.

## *2.4 Device construction and testing of epitaxial layers:*

We have studied the effect of MOCVD growth parameters on the properties of GaN and InGaN films. The $Al_2O_3(001)$ substrates were thermally cleaned under an $H_2$ flow. After that, a low temperature GaN buffer layer was grown to alleviate the lattice mismatch between the sapphire substrate and the GaN main layer. Then, the temperature was raised to the main layer growth temperature and was held for buffer layer annealing. The GaN main layer was then grown on the annealed buffer layer. For InGaN growth, the temperature was reduced and stabilized. $N_2$ was used as the carrier gas. Trimethylindium (TMIn) and trimethylgallium (TMGa) were used as the carrier gases in the reactor. The molar flowrate of TMGa, TMIn and the V/III molar ratio were fixed . A V/III molar ratio above 4300 was found to be needed to obtain a single crystal film.

*Effect of GaN Main Layer Growth Temperature:*

GaN films were grown with a fixed buffer layer temperature of 550 °C at various GaN main layer growth temperatures. Photoluminescence data showed a UV peak at 365 nm near band edge emission of wurtzite GaN with (bandgap = 3.4 eV). A broad yellow band was observed peaking around 550 nm corresponds to the energy of 2.2-2.3 eV. This band is caused by a transition from a shallow donor to a deep acceptor. The donor is thought to be a nitrogen vacancy and the deep acceptor is thought to be a complex consisting of a gallium vacancy and a carbon atom substituted for a nearest neighbor of the gallium site. The ratio of the area under the UV peak to the yellow band increases when the growth temperature is increased. This indicates that more defects are created at lower temperatures.

*Effect of GaN Buffer Layer Annealing Time:*

GaN films were grown without nitridation but with a fixed buffer layer temperature and at various annealing times, starting immediately after the temperature reaches the main layer growth temperature. The optimum conditions required to give the sharpest absorption band edge and flattest curve above band edge were found.

*Effect of GaN Buffer Layer Growth Temperature:*

GaN films were grown after buffer layer annealing with a fixed main layer temperature and at various GaN buffer layer growth temperatures, without prior nitridation. Hexagonal hillocks are observed by AFM measurements on the film with the buffer layer temperature of 540 and 550 °C. This indicates that such films are N-terminated. The buffer layer temperature of 575 °C and 590 °C, however, give a smooth morphology. The polarity of these smooth films was confirmed to be Ga-terminated. The smallest RMS roughness, found at 575 °C, was only 1.0 nm, which is preferable for subsequent film growth. The effective recombination lifetimes were determined by time-resolved PL measurements. The GaN film with the buffer layer temperature of 550 °C has more number of screw dislocations than the number of edge dislocations. In addition, these screw dislocations mostly run across the whole GaN film. The opposite behavior is found in the film with the buffer layer temperature of 575 °C.

*Effect of InGaN Growth Temperature:*

Our preliminary InGaN growth study using hydrogen as the carrier gas produced no significant indium incorporation in the film. Therefore, InGaN films were grown using nitrogen as the carrier gas at different growth temperatures on top of an approximately 550 nm thick GaN main layer previously deposited at 1080 °C. The buffer layer temperature was selected to be 575 °C because a smooth morphology is needed for an abrupt interface needed in heteroepitaxial growth. The indium content was determined and compared between XRD and RBS. The effect of biaxial strain in was taken into account by employing both symmetric and asymmetric XRD scan. In contrast, random RBS measurements are insensitive to strain and can be used to determine the indium content accurately. It was observed that as the growth temperature increases, the indium content decreases. Also, the indium content determined from XRD and RBS are comparable. The bandgap energies were determined to be between 2.12 EV to 3.21 eV, depending upon In content.

### *2.5 Future Plans:*

We will continue the MOCVD device development in order to first produce a GaN detector for UV emission detection, then grow an additional InGaN detector for addition detection of the 420 nm OSL emission from the $Al_2O_3$ dosimeters. Once the device is optimized, radiation testing will begin.

## *PUBLICATIONS & PRESENTATIONS RESULTING FROM THIS WORK:*

### *Publications*

1. E. G. Yukihara, R. Gaza, S. W. S. McKeever and C. G. Soares. *Optically stimulated luminescence and thermoluminescence efficiencies for high-energy heavy charged particle irradiation in* $Al_2O_3$*:C.* Radiat. Meas. **38**, 59-70 (2004).
2. E. G. Yukihara, V.H. Whitley, S. W. S. McKeever, A.E. Akselrod and M.S. Akselrod. *Effect of high doses and occupancy of deep traps on the optically stimulated luminescence of* $Al_2O_3$*:C.* Radiat. Meas. **38**, 317-330 (2004).

3 S.W.S. McKeever, M.W. Blair, E. Bulur, Razvan Gaza, Ramona Gaza, R. Kalchgruber, D.M. Klein and E.G. Yukihara. *Recent advances in dosimetry using the optically stimulated luminescence of $Al_2O_3$:C.* Radiat. Prot. Dosim. **109**, 269-276 (2004).

4. R. Gaza E.G. Yukihara and S.W.S. McKeever. *The response of thermally and optically stimulated luminescence from $Al_2O_3$:C to high-energy heavy charged particles.* Radiat. Meas. **38**, 417-420 (2004)
5. O. Goossens, F. Vanhavere, N. Leys, P. De Boever, D. O'Sullivan, D. Zhou, F. Spurny, E.G. Yukihara, R. Gaza and S.W.S. McKeever. *Radiation Dosimetry for Microbial Experiments in the International Space Station using Different Track-Etch and Luminescent Detectors.* Radiat. Prot. Dosim. (in press; 2005)
6. R. Gaza, E.G. Yukihara and S.W.S. McKeever. *The use of optically stimulated luminescence from $Al_2O_3$: C in the dosimetry of high-energy heavy charged particle fields.* Radiat. Prot. Dosim. (in press ; 2005).
7. E.G. Yukihara, and S.W.S. McKeever. *Ionization density dependence of the optically and thermally stimulated luminescence from $Al_2O_3$:C. (INVITED PAPER) Radiat. Prot. Dosim.* (in press, 2005).
8. R. Gaza, E. G. Yukihara, S.W.S. McKeever, O.Ávila, A.-E. Buenfil, I. Gamboa-deBuen, M. Rodríguez-Villafuerte, C. Ruiz-Trejo, and M.-E. Brandan. *Ionization density dependence of the optically stimulated luminescence dose response of $Al_2O_3$:C to low-energy charged particles.*Radiat. Prot. Dosim. (in press ; 2005).
9. T.-C. Chen, K. Poochinda and T. G. Stoebe. *Innovational Radiation Sensor by Integrating $Al_2O_3$:C Optically Stimulated Luminescent Dosimeter and GaN Detectors.* Radiat. Prot. Dosim. (in press, 2005).
10. E. G. Yukihara, R. Gaza, S. W. S. McKeever. *The potential of optically stimulated luminescence for dosimetry of astronauts.* Radiat. Res. (submitted, 2004).

***Presentations***

Solid State Dosimetry Conference, Yale, July 2004

1. O. Goossens, F. Vanhavere, N. Leys, P. De Boever, D. O'Sullivan, D. Zhou, F. Spurny, E.G. Yukihara, R. Gaza and S.W.S. McKeever. *Radiation Dosimetry for Microbial Experiments in the International Space Station using Different Track-Etch and Luminescent Detectors*)
2. R. Gaza, E.G. Yukihara and S.W.S. McKeever. *The use of optically stimulated luminescence from $Al_2O_3$:C in the dosimetry of high-energy heavy charged particle fields.*
3. E. G. Yukihara and S.W.S. McKeever. *Ionization density dependence of the optically and thermally stimulated luminescence from $Al_2O_3$:C.*
4. R. Gaza, E. G. Yukihara, S.W.S. McKeever, O.Ávila, A.-E. Buenfil, I. Gamboa-deBuen, M. Rodríguez-Villafuerte, C. Ruiz-Trejo, and M.-E. Brandan. *Ionization density dependence of the optically.*
5. T.-C. Chen, K. Poochinda and T. G. Stoebe. *Innovational Radiation Sensor by Integrating $Al_2O_3$:C Optically Stimulated Luminescent Dosimeter and GaN Detectors.*

LUMDETR Conf., Prague, September 2003

1. R. Gaza E.G. Yukihara and S.W.S. McKeever. *The response of thermally and optically stimulated luminescence from $Al_2O_3$:C to high-energy heavy charged particles*

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1. Ramona Gaza, Eduardo Yukihara and Stephen McKeever. *The potential of using optically stimulated luminescence from Al2O3:C dosimeters for space dosimetry: Behavior to HCP irradiations.* (Paris, 2002).
2. R. Gaza, E. G. Yukihara and S.W.S. McKeever. *The Optically Stimulated Luminescence of $Al_2O_3$:C to Heavy Charged Particles - Preliminary Results from 4th ICCHIBAN.* (Berkeley, 2003).
3. R. Gaza, O Goosens, S.W.S. McKeever, E.G. Yukihara and F. Vandehavre. Results from recent ground-based (HIMAC) and space-based (ISS) exposures using TL and OSL dosimeters (Vienna, 2004).

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1. E. G. Yukihara, R. Gaza, S. W. S. McKeever. The Potential of Optically Stimulated Luminescence as a Personal Dosimetry Method for Astronauts.

***Thesis***

1. R. Gaza. *Space Radiation Dosimetry: An OpticallY Stimulated Luminescence RadiatioN detector for Low-Earth Orbit.* Ph.D. Thesis, Oklahoma State University (Jluy, 2004).

***Patents***

1. T.C. Chen, T.G. Stoebe and S.W.S. McKeever. *Advanced Luminescent Radiation Sensor.* US Provisional Patent, (2004).