

**DEVELOPMENT OF SCINTILLATION GAMMA
RAY SPECTROMETERS FOR PLANETARY
MISSIONS AND METHODOLOGICAL
INVESTIGATIONS OF INFRARED STIMULATED
LUMINESCENCE SIGNALS FROM FELDSPARS**

*A thesis submitted to
Sardar Patel University, Vallabh Vidyanagar,
For the award of the degree of
Doctor of Philosophy
In
Physics*

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**THE DEGREE OF DOCTOR OF PHILOSOPHY
IN
PHYSICS**

by

Dipak Kumar Panda


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CERTIFICATE

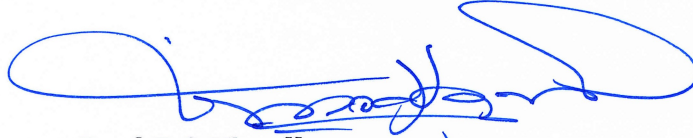
I hereby declare that the work presented in this thesis titled “Development of Scintillation Gamma Ray Spectrometers for Planetary Missions and Methodological Investigations of Infrared Stimulated Luminescence Signals from Feldspars” is original and has not formed the basis for the award of any degree or diploma by any University or Institute. The material obtained from other sources and used in the thesis have been acknowledged appropriately.


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Abstract

Chemical composition of various solar system objects such as planets, satellites and asteroids provides important clues towards understanding of their origin and evolution. The elemental abundances of such objects are established very early in the history of the solar system during the accretion of solar system material leading to their formation. Some of these objects also experienced subsequent thermal differentiation leading to formation of core, mantle and crust as in the case of the Earth. The chemical composition of a planetary surface allows us to infer whether accretionary process itself led to partial melting and caused large-scale redistribution of elements. Partial or complete melting will lead to a planetary surface that will be relatively depleted in Fe and Mg and enriched in O, Si, Al, and also in Na, K, Th and U. Partial losses of volatile elements such as C, N and S may also be inferred from surface composition.

Atmosphereless Planets or those with thin atmospheres (e.g. Mars, where surface pressure is ~ 7 millibars) are continuously irradiated with galactic cosmic rays (GCR), mainly protons and alpha particles with energies typically $\sim \text{GeV/nucleon}$. These GCR particles enter the planetary surface and produce a cascade of secondary particles, including ~ 7 neutrons (for the case of Moon) with energies of $\sim 0.1\text{--}20$ MeV, per primary particle. Many of these secondary neutrons can produce gamma rays in inelastic scattering ($n, x\gamma$) reactions, where x is usually a neutron. Neutrons with energies below the first excited level of target nuclei in planetary surface can be elastically scattered by nuclei, escape from the surface (about one third of neutrons), or be captured by nuclei in the (n, γ) reaction when the neutron energy is of the order of ~ 0.025 eV. Both the inelastic and the neutron capture gamma ray lines are those classically used in planetary applications. Gamma rays can also be produced by the decay of naturally available radioactive elements such as K, U, Th. In comparison to X-ray fluorescence spectrometry, compositional data from gamma spectrometry is more representative of the planetary sub-surface as gamma rays come from

depths of a few centimeters to tens of centimeters whereas characteristic X-rays have a maximum interaction depth of the order of tens of microns.

Three types of detector systems are suitable for 0.1 to 10 MeV energy range. The scintillation detectors such as NaI(Tl), CsI(Tl) and Bismuth Germanate (BGO) have been used in many space missions for gamma ray spectroscopy. Their advantages include heritage, easy availability, high detection efficiency, and easily realizable large area detectors. The main disadvantage of these detectors is their poor energy resolution, and are not considered for the present experiment. High Purity Germanium (HPGe) detectors have been used in the 2001 Mars Odyssey Mission, Selene (Kaguya) moon mission and Messenger Mercury mission. These detectors have significantly better spectral resolution than the scintillation detectors used on previous missions. The GRS instrument on Odyssey used a passively cooled (~ 85 K) high-purity n-type germanium crystal that detects gamma rays in the energy range of 0.1 to 10 MeV. However, the HPGe detectors need to be cooled to liquid nitrogen temperatures (77 K) to achieve high energy resolution levels, implying the use of a Stirling cycle cooler or a passive cooling system, require high power (~ 50 W) and a long time for development.

The newly developed cerium doped lanthanum bromide ($\text{LaBr}_3\text{:Ce}$) crystal is the latest among the family of the scintillation counters, and has an advantage over conventional room temperature detectors. It has a high effective atomic number, high light yield, and therefore, the energy resolution and detection efficiency of $\text{LaBr}_3\text{:Ce}$ detector is superior to NaI(Tl). The energy resolution of this detector is 2.8% at 662 keV (^{137}Cs) and $\sim 1.6\%$ at 2615 keV (^{208}Tl). The light output of this scintillator is 7 times higher in comparison to BGO, and 1.6 times higher compared to NaI(Tl). Furthermore, this detector does not require active or passive cooling systems as required for HPGe detectors, and can be operated at room temperature.

In this thesis, a $\text{LaBr}_3\text{:Ce}$ gamma ray spectrometer has been developed for a future planetary orbiter mission with the primary objective of determining the

abundance and distribution of Th, U, K, and other major elements on the entire planetary surface by measuring gamma ray signals produced by radioactive decay, neutron inelastic scattering and neutron capture reactions in the energy region 0.03 to 8 MeV. Based on weight, power, and operating temperature considerations for the spacecraft payloads, a LaBr₃:Ce gamma detector would appear be the best choice for the gamma spectrometer and was our original choice for the proposed gamma ray spectrometer. However, the intrinsic activity count-rate for a 3"× 3" LaBr₃:Ce gamma ray spectrometer observed to be ~61 counts s⁻¹ (i.e. ~0.18 counts s⁻¹ cm⁻³) for the ⁴⁰K energy window (1400-1520 keV) and ~3.4 counts s⁻¹ for the ²³²Th (2550-2700 keV) energy window (Panda et. al. 2016). Thus, the gamma ray background arising from intrinsic activity of ¹³⁸La and due to the contamination of ²²⁷Ac are significantly large and makes it difficult to make abundance measurements of Th, U and K in the 0.05-3 MeV energy region.

In light of the above, we focused on the development of a CeBr₃ gamma ray spectrometer with in-house developed electronics and software. The energy resolution of the CeBr₃ gamma ray spectrometer using front-end and processing electronics developed in-house has been measured at 662 and 1274 keV to be 4.0% and 2.8% respectively. The intrinsic activity count-rate for the 1"× 1"CeBr₃ gamma ray spectrometer is ~0.03 counts s⁻¹ for the ⁴⁰K energy window (1400-1520 keV), and ~0.001 counts s⁻¹ for the ²³²Th (2550-2700 keV) energy window. The U concentration of a sample (3A) from a granite rock was estimated to be ~2.1 ppm and agrees with the 2.04 ppm value determined using a HPGe gamma ray spectrometer. The K concentration of sample 3A was estimated to be 3.7%, and is consistent with the 3.8% value determined independently using a HPGe gamma ray spectrometer.

The third section of the thesis is focused on the determination of luminescence ages of feldspar grains since the luminescence signal from feldspar has potential for measuring large doses (~kGy). Luminescence technique is used to evaluate the time that has elapsed since the mineral grains crystalized, were exposed to daylight or heated beyond 400°C. These methods use optically and

thermally sensitive light signal from minerals such as quartz and feldspar. We investigate the source of the IRSL signal in two feldspars obtained from NIST, viz., Standard Reference Material (SRM) 99b (soda feldspar) and K-feldspar SRM 607. For SRM 607, the TL glow curve consists of a several peaks at $\sim 95^{\circ}\text{C}$, 160°C , 260°C and 340°C . In the soda feldspar SRM 99b, a broad TL peak is observed at temperatures between 140°C - 190°C ; this TL signal is absent in the preheated SRM 99b aliquot, and a TL peak at $\sim 300^{\circ}\text{C}$ is observed in the glow curve. Although IRSL signals from the soda-feldspar SRM 99b indicate no significant depletion at temperatures below 200°C , the pulse anneal data for the K-feldspar sample SRM 607 does show a 30% loss in IRSL while heating upto 200°C . For the natural feldspar extracts considered here (SUN-1-GR and SHRD-1), the equivalent dose value remains constant for preheat temperatures between 80°C - 320°C . It has been previously argued that a single trap (around 410°C) may be primarily responsible for the IRSL signal (Murray et al. 2009). The preheat plateaus from natural feldspar extracts presented here support the conclusion that low temperature TL peaks do not make a significant contribution to IRSL in several feldspar samples.

DECLARATION

I hereby declare that the work incorporated in the present thesis entitled, “Development of Scintillation Gamma Ray Spectrometers for Planetary Missions and Methodological Investigations of Infrared Stimulated Luminescence Signals from Feldspars” is my own work and is original. This work (in part or in full) has not been submitted to any University for the award of a Degree or a Diploma.

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Figure.5.1: Explanation of luminescence (TL, OSL) processes in term of the band theory of solids. The symbol L and T indicate hole traps and electron traps respectively. E denotes the activation energy, which for mineral such as quartz, K-feldspars range from 0.7 – 2 eV.

Figure.5.2: Energy level representation of the IRSL process (Hutt et. al. 1988).

Figure.5.3: Sketch of the RISO TL/OSL reader.

Figure 5.4: TL glow curves for SRM 607, SRM 99b and SHRD-1 obtained using a heating rate of 2°C s^{-1} .

Figure 5.5: Variation of the IRSL signal with preheat temperature (plus annealing curves) for SRM 607 and SRM 99b. The IRSL are normalized with respect to their initial value.

Figure 5.6: TL glow curves for SRM 607 measured after infrared stimulation at 60°C for different durations.

Figure 5.7a: TL lost due to IR exposure for SRM 607 and SRM 99b, measured after preheating at 250°C for 60 seconds and inferred stimulation.

Figure 5.7b: TL lost due to IR exposure for SRM 99b, measured after preheating at 250°C for 60 seconds and inferred stimulation.

Figure 5.8: TL lost due to IR exposure for SRM 607 and SRM 99b, measured after preheating at 320°C for 60 seconds and inferred stimulation.

Figure 5.9: Variation of equivalent dose with preheat temperature for SHRD-1 and SUN1GR.

List of Publications:

1. Panda, D. K., Banerjee, D., Goyal, S. K., Patel, A. R. and Shukla, A. D., "Development of a Cerium bromide gamma ray spectrometer for space applications." *Advances in Space Research*, 2017, 60, 1307-1314, doi.org/10.1016/j.asr.2017.06.016.
2. Panda, D. K., Banerjee D., Goyal S. K., Patel A. R., Shukla, A. D., "Development of a Cerium-doped Lanthanum Bromide gamma-ray spectrometer for planetary missions and feasibility studies for determination of elemental abundances of radioactive elements (Th, K and U).", *Current Science*, 2016, 110, 2135-2138.

Conference Proceedings:

1. Development of LaBr₃:Ce and CeBr₃ gamma ray spectrometers for space applications.
Dipak K. Panda, D. Banerjee, S.K. Goyal, A.R Patel, and A.D. Shukla, SSD-18, 4th -8th July-2016.
2. On the application of visible - NIR reflectance spectroscopy for distinguishing feldspar and quartz OSL signals
Debabrata Banerjee, Dipak Kumar Panda, SSD-18, 4th -8th July-2016.
3. New Results from laboratory model of a LaBr₃:Ce based gamma ray spectrometer.
D. K. Panda, D. Banerjee, S.K. Goyal, A. R. Patel and Tinkal Ladiya, NSSS, 9-12th Feb-2016.
4. A LaBr₃:Ce Gamma Ray Spectrometer for Future Planetary Mission.
D. K. Panda, D. Banerjee, S.K. Goyal, A. R. Patel and Tinkal Ladiya, NSSS, 29th Jan- 1st Feb 2014.

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