## 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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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  $\sim$ 7 millibars) are continuously irradiated with galactic cosmic rays (GCR), mainly protons and alpha particles with energies typically ~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  $\sim 0.1$ -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  $(n,\gamma)$  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 ( $\sim$ 50W) and a long time for development.

The newly developed cerium doped lanthanum bromide (LaBr<sub>3</sub>: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 LaBr<sub>3</sub>:Ce detector is superior to NaI(Tl). The energy resolution of this detector is 2.8% at 662 keV (<sup>137</sup>Cs) and ~1.6% at 2615 keV (<sup>208</sup>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 LaBr<sub>3</sub>: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 LaBr<sub>3</sub>: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  $3'' \times 3''$  LaBr<sub>3</sub>:Ce gamma ray spectrometer observed to be ~61 counts s<sup>-1</sup> (i.e. ~0.18 counts s<sup>-1</sup> cm<sup>-3</sup>) for the <sup>40</sup>K energy window (1400-1520 keV) and ~3.4 counts s<sup>-1</sup> for the <sup>232</sup>Th (2550-2700 keV) energy window (Panda et. al. 2016). Thus, the gamma ray background arising from intrinsic activity of <sup>138</sup>La and due to the contamination of <sup>227</sup>Ac 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 CeBr<sub>3</sub> gamma ray spectrometer with in-house developed electronics and software. The energy resolution of the CeBr<sub>3</sub> 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  $1"\times 1"$ CeBr<sub>3</sub> gamma ray spectrometer is ~0.03 counts s<sup>-1</sup> for the <sup>40</sup>K energy window (1400-1520 keV), and ~0.001 counts s<sup>-1</sup> for the <sup>232</sup>Th (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 ( $\sim$ 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 ~95°C, 160°C, 260°C and 340°C. In the soda feldspar SRM 99b, a broad TL peak is observed at temperatures between 140°-190°C; this TL signal is absent in the preheated SRM 99b aliquot, and a TL peak at  $\sim$ 300°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.

Dipak Kumar Panda

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## **Table of Contents**

Acknowledgements				
List	of Tab	oles	х	
List of Figures			xii	
1.	Introduction			
	1.1	Methods used for Planetary Exploration	3	
	1.2	Production of Gamma Ray on Planetary Surface	5	
		1.2.1 Gamma Ray Production by Galactic Cosmic Rays	5	
		1.2.2 Gamma Ray Production by Decay of Natural Radioactive		
		Elements	7	
	1.3	GRS Results from Previous Planetary Missions	7	
		1.3.1 Gamma Ray Experiment on Moon	7	
		Apollo Mission	7	
		Lunar Prospector Mission	8	
		Selene (Kaguya) Mission	9	
		Chang'E Mission	11	
		1.3.2 Gamma Ray Experiment on Mars	12	
		1.3.3 Gamma ray Experiments on Mercury	13	
		1.3.4 Gamma Ray Experiments on Dawn and Ceres (Asteroids)	14	
	1.4	Motivation of the Present Thesis	15	
	1.5	Research Objectives of Present Study	17	
	1.6	Scope of the Present Thesis	17	

		-y -p	
Techniques 2			
2.1	Princi	iple	20
	2.1.1	Radioactive Decay	20
	2.1.2	Gamma Decay	22
2.2	Intera	action of Gamma Rays with Matter	22
	2.2.1	Photoelectric Absorption	22
	2.2.2	Compton Scattering	23
	2.2.3	Pair Production	25
	2.2.4	Total Absorption	25
2.3	Radia	tion Detector	26
	2.3.2	Types of Radiation Detector	26
2.4	Chara	cteristics of Gamma Ray Spectrometer	28
	2.4.1	Detector Response	29
	2.4.2	Energy Resolution	31
	2.4.3	Detection Efficiency	33
		Absolute Efficiency	33
		Intrinsic Efficiency	34
		Photopeak Efficiency	34
2.5	Analy	sis of Pulse Height spectra	34
2.6	Curre	nt Development in Scintillation Crystals	36

### 2. Gamma Ray Spectrometry Principles and Detection

	2.7	Simul	Simulation of Performance of Scintillation Detector		
		2.7.1	GEANT4 Tool Kit	37	
		2.7.2	Detector Geometry	38	
		2.7.3	Efficiency of LaBr3:Ce, CeBr3 and NaI(Tl) detector	39	
		2.7.4	Simulation of Response of LaBr3:Ce from Radioactive Sources	43	
3.	Dev	elopm	ent of a Cerium Doped Lanthanum Bromide		
	Gam	ima Ra	ay Spectrometer for Planetary Missions and		
	Feas	sibility	v Studies	46-91	
	3.1	Introd	duction	46	
	3.2	Ceriu	m Doped Lanthanum Bromide Detector	47	
		3.2.1	Physical properties of LaBr3:Ce detector	48	
		3.2.2	Mechanism of Scintillation	49	
		3.2.3	Intrinsic Activity of LaBr3:Ce	50	
	3.3	Feasil	bility Studies of LaBr3:Ce Using Commercial Setup	51	
		3.3.1	Calibration & Energy resolution	52	
	3.4 Development of gamma ray spectrometer		opment of gamma ray spectrometer	56	
		3.4.1	Detector Configuration	58	
		3.4.2	Front End Electronics	59	
			3.4.2.1 Charge sensitive pre-amplifier (CSPA)	59	
			3.4.2.2 Shaping amplifier	60	

	3.4.3	4.3 Processing Electronics		
		3.4.3.1 Event Triggering	65	
		3.4.3.2 Peak Detector	65	
		3.4.3.3 Analogue to Digital Converter (ADC)	68	
		3.4.3.4 FPGA based data acquisition system	69	
	3.4.4.	Data readout system using LabVIEW	70	
3.5	3.5 Results			
	3.5.1	Experimental setup	72	
	3.5.2	Calibration and Energy resolution	73	
	3.5.3	Background measurement and Intrinsic Activity	80	
	3.5.4	Estimation of K, U, and Th Concentration in		
		Terrestrial Samples	84	
	3.5.5	Comparative Study of LaBr3:Ce and NaI(Tl)	86	
	_			
Cerium Bromide Gamma Ray Spectrometer for Space				
Applications 92-			92-124	
4.1	Introduction			
4.2	Cerium Bromide Detector (CeBr <sub>3</sub> )			
	4.2.1	Mechanism of Scintillation of CeBr <sub>3</sub>	94	

4.

## 4.2.2 Characteristics of CeBr<sub>3</sub> 95

•	Decay time	95

• Light output and energy resolution 95

		Intrinsic activity	95
4.3		Feasibility study of $CeBr_3$ for Space application	96
		4.3.1 Experimental Setup	96
		4.3.2 Shaping Amplifier and Discriminator	99
		4.3.3 Peak Detector and Analog-to-Digital Converter	100
		4.3.4 FPGA Processing Electronics and Data Acquisition	101
		4.3.5 Calibration	102
		4.3.6 Energy Resolution	104
		4.3.7 Count Rate Variation with Change in Source to Detector Distance for $CeBr_3$	107
		4.3.8. Response of $CeBr_3$ with Biasing Voltage	109
		4.3.9 Shaping time vs energy resolution	112
		4.3.10. Intrinsic Activity of CeBr <sub>3</sub> Detector	114
	4.4	Comparison of CeBr3 with LaBr3:Ce and NaI(Tl)	116
	4.6	Estimation of K, U and Th concentration from Geological Sample	121
5.	Meth	odological Investigations of Infrared Stimulated	
	Luminescence Signals from Feldspars125-2		
	5.1	Introduction	125
	5.2	Luminescence	126
	5.3	Thermoluminescence dating	127
		5.3.1 Annual dose determination	128

Bibliography 15			150-165
6	Conc	lusion and Future Work	146-149
	5.11	Discussion	142
		5.10.2 Effect of IR preheating at elevated temperature	139
		5.10.1 IR stimulation and TL measurements without preheat	138
	5.10	Pulse Annealing Experiments	136
	5.9	Luminescence Glow Curve	134
	5.8	Experimental Procedure & Sample	134
	5.7	Sample Preparation Methodology	133
	5.6	Samples	132
	5.5	Infrared Luminescence Signals from Feldspar	131
	5.4	Infra-Red Stimulated Luminescence Dating	129

### **List of Tables**

#### **Chapter 2**

Table 2.1. Comparison of efficiency for 3" x 3" scintillation detectors.

#### **Chapter 3**

Table 3.1: Comparison of properties of gamma ray detectors.

- Table 3. 2: Properties of LaBr<sub>3</sub>:Ce.
- Table 3.3: Properties of PMT coupled to LaBr<sub>3</sub>:Ce detector.
- Table 3.4: Comparison of energy resolution achieved by GRS developed with Commercial electronics and in-house.
- Table 3.5: Comparison of intrinsic activity measured from GRS developed withcommercial electronics module & the module developed in house.

Table 3.6: Concentration of US#110 & BBS#2 measured with HPGe.

- Table 3.7: Comparison of Intrinsic activity from LaBr<sub>3</sub>:Ce & NaI(Tl).
- Table 3.8: Elemental Concentrations of samples determined using HPGe and NaI(Tl) gamma ray spectroscopy.

#### **Chapter 4**

- Table 4.1: Properties of CeBr<sub>3</sub>.
- Table 4.2: The dependence of count rate on source to detector distance.
- Table 4.3: Intrinsic activity of CeBr<sub>3</sub> at different energy range.
- Table 4.4: Comparison of intrinsic activity measure with LaBr<sub>3</sub>:Ce, CeBr<sub>3</sub> & NaI(Tl) at different energy windows.

### Chapter 5

Table 5.1: U, Th and K concentration measured using Gamma Ray Spectrometer.

Table 5.2: OSL age measured using quartz mineral.

Table 5.3: IRSL age measured using feldspar mineral.

#### **List of Figures**

#### **Chapter 1**

- Figure 1.1: Solar System formation from Interstellar Molecular Cloud (Adopted from Pearson Prentice Hall, Inc, 2005).
- Figure 1.2: Production of gamma rays on a Planetary Surface.

#### **Chapter 2**

- Figure 2.1: Schematic representation of Photoelectric absorption process.
- Figure 2.2: Schematic representation of Compton scattering process.
- Figure 2.3: Relative importance of tree types of gamma ray interaction.
- Figure 2.4: Schematic of gas filled detector.
- Figure 2.5: Schematic semiconductor detector.
- Figure 2.6: Representation of the gamma detector spectrum from a mono-energetic gamma source.
- Figure 2.7: Definition of detector energy resolution.
- Figure 2.8: Pulse height distribution of gamma ray spectrum.
- Figure 2.9: Construction of 3" x 3" detection with Geant4 simulation with 0.5 mm Al casing.
- Figure 2.10: Interaction of beam with LaBr<sub>3</sub>:Ce Detector.
- Figure 2.11: Photo-peak efficiency simulated for different size of LaBr<sub>3</sub>:Ce detector.
- Figure 2.12: Photo-peak efficiency simulated for different size of CeBr<sub>3</sub> detector.

#### Figure 2.13: Photo-peak efficiency simulated for different size of NaI(Tl) detector.

- Figure 2.14: Comparison of photo-peak efficiency simulated for 3" x 3" LaBr<sub>3</sub>:Ce, CeBr<sub>3</sub> and NaI(Tl) detector.
- Figure 2.15: Response of a 3" x 3" LaBr<sub>3</sub>:Ce detector when exposed to radioactive sources. (a) <sup>22</sup>Na, (b) <sup>60</sup>Co and (c) <sup>137</sup>Cs.

#### **Chapter 3**

- Figure 3.1: Decay scheme of <sup>138</sup>La.
- Figure 3.2: Self-counting background spectrum of LaBr<sub>3</sub>:Ce detector.
- Figure 3.3: Response of LaBr<sub>3</sub>:Ce to different radio-active sources, (a) <sup>214</sup>Am, (b) <sup>133</sup>Ba, (c) <sup>137</sup>Cs, (d) <sup>22</sup>Na, and (e) <sup>152</sup>Eu.
- Figure 3.4: Linear curve of ADC Channel vs Energy for LaBr<sub>3</sub>:Ce.
- Figure 3.5: Energy resolution achieved at different energies for LaBr3:Ce detector.
- Figure 3.6: Block diagram of LaBr<sub>3</sub>:Ce gamma ray spectrometer.
- Figure 3.7a: 3" x 3 " LaBr3:Ce detector couple with a 3.5" XP-5700 PMT.
- Figure 3.7b: Front view of voltage divider & pre-amplifier.
- Figure 3.8: Snapshot of pre-amplifier output of LaBr<sub>3</sub>:Ce, captured with Tektronix Oscilloscope detector.
- Figure 3.9: Schematic of CR-RC Network.
- Figure 3.10: The time and frequency domain response of CR, RC and CR-RC.
- Figure 3.11: Block schematic of shaping amplifier.
- Figure 3.12: Snapshots of different stage of shaping amplifier output captured with Tektronix Oscilloscope.
- Figure 3.13: Output at different stage of shaping amplifier.

Figure 3.14: Block schematic of peak detection.

- Figure 3.15: Bock schematic of PH-300 peak detector, (adopted from Amptek data sheet).
- Figure 3.16: Different mode of operation of PH-300 (adopted from Amptek data sheet).
- Figure 3.17: Block diagram of interfacing of different control signals from FPGA to Peak detector, ADC and interfacing DAQ (data acquisition) card.
- Figure 3.18: Breadboard module of processing electronics for gamma ray spectrometer.
- Figure 3.19: Snapshot of data acquisition software developed using LabVIEW.
- Figure 3.20: Timing diagram for data processing of gamma ray spectrometer.
- Figure 3.21: Laboratory Experimental setup.
- Figure 3.22: Calibration of LaBr<sub>3</sub>:Ce using <sup>22</sup>Na radioactive source.
- Figure 3.23: Calibration curve obtained using <sup>22</sup>Na radioactive source and internal activity of LaBr<sub>3</sub>:Ce.
- Figure 3.24: Calibration curve obtained using different radioactive source.
- Figure 3.25: Gamma ray spectra measured with LaBr<sub>3</sub>:Ce using different radio-active source. (a) <sup>241</sup>Am, (b) <sup>133</sup>ba, (c) <sup>137</sup>Cs, (d) <sup>22</sup>Na and <sup>152</sup>Eu.
- Figure 3.26: Energy resolutions with varying energy for LaBr<sub>3</sub>:Ce detector.
- Figure 3.27a: Gamma ray background spectra obtained using LaBr<sub>3</sub>:Ce gamma ray spectrometer.
- Figure 3.27b: Background spectra of LaBr<sub>3</sub>:Ce measured upto 3 MeV. The plot shows peaks at 789 keV and 1436 keV intrinsic activity of <sup>138</sup>La.

- Figure 3.27c: Presence of intrinsic activity in the energy range of 1.7 3 MeV within LaBr<sub>3</sub>:Ce detector due to <sup>227</sup>Ac.
- Figure 3.28: Comparison of gamma ray spectrums measured with LaBr<sub>3</sub>:Ce from background and samples (a) US#110 and (b) BBS#2.
- Figure 3.29: Response of NaI(Tl) from <sup>22</sup>Na radioactive source.
- Figure 3.30: Calibration curve of NaI(Tl) detector.
- Figure 3.31: Background spectra measured with 2" x 2" NaI(Tl) detector.

Figure 3.32a: Gamma ray spectra from US#110 measured with NaI(Tl).

Figure 3.32b: Gamma ray spectra from BBS#2 measured with NaI(Tl).

#### **Chapter 4**

- Figure 4.1: X-ray emission of LaBr<sub>3</sub>:5%Ce and three different size of CeBr<sub>3</sub> (adopted from Quarati et. al. 2013).
- Figure 4.2: Comparison of self-activity for 1" x 1" CeBr<sub>3</sub>, LaBr<sub>3</sub>:Ce, LaCl<sub>3</sub>, and NaI(Tl) (Adopted from Gauss et. al. 2009).
- Figure 4.3: Front view of CeBr<sub>3</sub> scintillation detector.
- Figure 4.4: Block diagram of CeBr<sub>3</sub> gamma ray spectrometer.
- Figure. 4.5: Response of different radioactive sources measured with CeBr3 detector.
- Figure 4.6: Calibration curve of CeBr<sub>3</sub>.
- Figure 4.7: Response of radioactive sources measurement with CeBr<sub>3</sub>. (a) <sup>241</sup>Am, (b) <sup>133</sup>Ba, (c) <sup>137</sup>Cs, (d) <sup>22</sup>Na, and (e) <sup>152</sup>Eu.
- Figure 4.8: Variation of energy resolution with energy for CeBr<sub>3</sub>.
- Figure 4.9: Variation of photo-peak count rate expressed as a function of source to detector distance in side Pb shield of 4" wall thick.

- Figure 4.10: Linearity for Biasing Voltage vs ADC Channel at 662 keV(<sup>137</sup>Cs).
- Figure 4.11: Plot for Biasing Voltage vs. Energy Resolution at 662 keV (<sup>137</sup>Cs).
- Figure. 4.12: Linearity curve for Energy vs ADC Channel at different Biasing Voltage.
- Figure. 4.13: Plot for Energy resolution vs. Energy at different Biasing Voltage.
- Figure 4.14: (a) Spectral response of <sup>22</sup>Na radioactive source at different shaping time,(b) variation of ADC Channel with shaping time. (c) Energy resolution vs. shaping time.
- Figure 4.15a: Gamma ray background spectra measured using 1" x 1" CeBr<sub>3</sub> gamma ray spectrometer kept within a 4" thick lead shield. CeBr<sub>3</sub> detector.
- Figure 4.15b: Background measurement of CeBr<sub>3</sub> in the energy range 1.3 MeV to 2.1 MeV.
- Figure 4.16: Comparison of intrinsic activities from LaBr<sub>3</sub>:Ce, CeBr<sub>3</sub> and NaI(Tl).
- Figure 4.17: Comparison of response of radioactive source from LaBr<sub>3</sub>:Ce, CeBr<sub>3</sub> and NaI(Tl). (a)<sup>133</sup>Ba and (b)<sup>152</sup>Eu.
- Figure.4.18: Comparison of energy resolution vs energy correlation from LaBr<sub>3</sub>:Ce, CeBr<sub>3</sub> and NaI(Tl).
- Figure 4.19: Comparison of background spectra and K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub> and gamma ray spectra from KCl measured with CeBr<sub>3</sub> detector.

#### **Chapter 5**

- 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<sup>o</sup> 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<sup>o</sup> C for different durations.
- Figure 5.7a: TL lost due to IR exposure for SRM 607 and SRM 99b, measured after preheating at 250<sup>o</sup> 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<sup>o</sup> 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:**

- 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.
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## **Conference Proceedings:**

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

## **Chapter 1**

### **1.** Introduction

The solar system originated ~4.567 billion years ago, but the complete understanding of origin and evolution of the solar system is still awaited. It is widely believed that the solar system formed within 100 million years after the formation of the Sun, which itself took less than 1 million years. From astronomical observations, it has been observed that stars are not born in isolation, but in clusters of our galaxy (Montmerle et. al. 2006). Stars are formed in the densest regions of the interstellar medium (ISM), called molecular clouds. The ISM is mainly filled with gas and dust which exists between the stars within a galaxy.

The most widely accepted nebular hypothesis for the formation of our solar system suggests that the gravitational collapse of a giant molecular cloud fragment led to the formation of the proto-sun at its centre and a rotating disk of gas and dust, called the solar-nebula, surrounding the nascent sun (Bhattacharya & Litchman, 2017), as shown in Figure 1.1. The solar system objects (planets, satellites, comets, and asteroids) formed out of this nebula. Planets are byproducts of star formation and are formed from the solar nebula, the disk-shaped cloud of gas and dust left over from the Sun's formation (Goldreich & Ward, 1973). When a star forms from a rotating molecular cloud, the angular momentum conservation law forces this collapsing cloud matter to form a rotating accretion disk around the newly forming star. Initially, disks rapidly accrete material onto the star but, as the surrounding molecular core is used up or disperses, the accretion rate decreases and a small amount of material persists. The disk, in this late stage, can be called as a protoplanetary disk. (Williams & Cieza, 2011). The dust grains in the protoplanetary disk clustered to form meter size bodies, which in turn collide to form larger bodies (~kilometeres), known as planetesimals and subsequently they grow to become planets.



Figure 1.1: Solar System formation from Interstellar Molecular Cloud (Adopted from Pearson Prentice Hall, Inc, 2005).

The protoplanetary disks exhibit a range of temperatures, hot near the star, cooler farther away. The inner solar system was too hot for volatile molecules (H<sub>2</sub>O, methane etc.) to condense so that the planetesimals that were created from higher melting materials such as iron, nickel, Aluminium etc. and rocky silicates. These rocky bodies comprise the terrestrial planets (Mercury, Venus, Earth and Mars). The gas giant (Jupiter, Saturn, Uranus and Neptune) formed beyond the frost line (3.1 AU, Martin and Livio, 2012), the line between Mars and Jupiter. These regions are sufficiently cool for volatile icy compounds to remain solid. The terrestrial planets consitutes only 0.6% of the mass nebula, while the giant planets <99% of all the mass orbiting sun.

#### **1.1 Methods used for Planetary Exploration**

The Chemical composition of various solar system objects such as planets, satellites, and asteroids provides important clues towards understanding 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 the 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 the 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 the surface composition.

Elemental composition of a planetary surface can be deduced from in-situ measurements or by remote sensing technique and laboratory analysis of returned samples. These techniques have been applied in the case of the Moon, while the study of other planets, satellites, and asteroids have been carried out using remote sensing technique with the exception of Mars where in-situ measurements were also carried out. X-ray fluorescence, gamma ray, alpha ray, neutron and neutral atom spectroscopy are some of the basic tools for mapping the planetary surfaces. High energy (>100 keV) gamma ray spectroscopy is an

important technique for remote sensing studies of the chemical composition of planetary surfaces and has been used to study the surface composition of the Moon, Mars, and Asteroids at various spatial resolutions.

In the case of moon, a LaBr<sub>3</sub>:Ce (Ce doped Lanthanum Bromide) gamma ray spectrometer was initially proposed for an orbiter mission with the primary objective of determining the abundance and distribution of Th, U, Mg, K, Ti, Ca, Al, Si, Fe, and O 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. Iron is a key element, along with the Fe to Mg abundance ratio, to test various hypotheses regarding the origin of the moon (Lucey et al. 1995). The formation of the ancient lunar crust requires flotation of plagioclase feldspar in a global magma ocean, implying that the lunar crust has Al-rich and Fe-poor composition (Warren, 1990). Models of magma ocean evolution suggest that the Fe content should be less than 4% (by weight) in anorthositic rocks that formed from plagioclase assemblages which floated to the top of the magma ocean (Warren, 1990).

For Mars, the knowledge of dust chemistry is incomplete, and it is not known to what extent the source rock composition of the fine-grained Martian dust is representative of Martian surface rocks. A comparison of the global average of the dust composition and the composition of the Martian rocks is necessary for understanding the weathering processes on Mars. The abundances of Cl in the Martian soil and caliche like deposits such as duricrust can be useful to investigate the presence of NaCl and to correlate the salt concentration with other geological indicators of the action of water. It will be possible to map the composition of Martian geological units which record a history of volcanism on Mars and to look for compositional changes with time.

The recently developed cerium doped lanthanum bromide (LaBr<sub>3</sub>: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 LaBr<sub>3</sub>:Ce detector is superior to NaI(Tl). The energy resolution of this detector is 2.8% at 662 keV ( $^{137}$ Cs) and ~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 (High Purity Germanium) detectors and can be operated at room temperature. Our initial development efforts used a 3-inch diameter, 3-inch long LaBr<sub>3</sub>:Ce crystal optically coupled to a 3.5-inch diameter photomultiplier tube(PMT). The output of the PMT will be fed into charge sensitive pre-amplifier, which converts the charge into a voltage pulse. The outputs of the preamplifiers will be further shaped and amplified by a linear amplifier and then processed by a single 14-bit analog to digital converter (ADC). These data will be sent to FPGA (Field Programmable Gate Array) based processing electronics for further processing and a LabVIEW based software will be used to acquire data.

#### **1.2 Production of Gamma Ray on Planetary Surface**

#### **1.2.1 Gamma Ray Production by Galactic Cosmic Rays**

The planetary surface is continuously irradiated with solar and galactic cosmic rays (GCR). The GCR are mainly composed of protons and alpha particles with energies typically ~GeV/nucleon. The GCR are mainly comprised of ~ 87% protons (nuclei of <sup>1</sup>H), ~12% alpha particles (nuclei of <sup>4</sup>He), and ~1% heavier nuclei. The GCR arrive in the solar system with energies typically of ~0.1–10 GeV/nucleon. (Reedy 1972).

For energies of several GeV and less, the effect of the solar magnetic fields on the GCR energy spectrum is very significant and the number of low-energy GCR particles decreases with decreasing distance from the sun. This radial modulation of the GCR is an important factor in comparing the rates of nuclear reactions at different radial distances from the sun. This effect has been seen in meteorites whose orbits extended several AU (Astronomical unit) beyond the earth's orbit (Forman et al., 1971; Fireman and Spannagel 1971)



#### Figure 1.2: Production of gamma rays on a Planetary Surface.

These GCR particles enter the planetary surface and produce a cascade of secondary particles, including  $\sim$ 7 neutrons for the case of the moon while  $\sim$ 10 neutrons for the case of Mars, with energies of  $\sim 0.1-20$  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 the planetary surface can be elastically scattered by nuclei, escape from the surface of planet (about one third of neutrons), or be captured by nuclei in the  $(n, \gamma)$  reaction when the neutron energy is of the order of  $\sim 0.025$  eV. Gamma rays can also be produced by the decay of radionuclides produced by such cosmic ray-induced nuclear reactions. The gamma rays, thus produced, are transported within the planetary surface; some of them are absorbed, while others can induce pair production, or are scattered out of the planetary surface. Both the inelastic and the neutron capture gamma ray lines are those classically used in planetary applications. In comparison to X-ray fluorescence spectrometry, compositional data from gamma spectrometry is more representative of the planet 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.

# **1.2.2 Gamma-Ray Production by Decay of Natural Radioactive Elements**

Radioactive decay of naturally available radioactive elements such as K, U, Th, and Rb comprise another source of gamma rays. In natural Potassium (<sup>40</sup>K), 0.012% of radioactive <sup>40</sup>K is present; the radioactive <sup>40</sup>K decays to the excited level of <sup>40</sup>Ar at an energy of 1.461 MeV. This excited <sup>40</sup>Ar almost immediately decays to the <sup>40</sup>Ar ground state by emitting a 1.461 MeV gamma ray. Other naturally radioactive elements like <sup>232</sup>Th (half-life 14 Ga), <sup>238</sup>U (4.5 Ga), and <sup>235</sup>U (0.70 Ga) can be also be used for determining elemental abundances of Th and U by estimation of signals arising from gamma rays from isotopes in the respective decay series. Several isotopes are produced by <sup>232</sup>Th and <sup>238</sup>U decay series and emit gamma rays which can be used for producing elemental maps. Thorium is usually mapped using the 2.615 MeV gamma ray emitted by its <sup>208</sup>Tl daughter. Other strong gamma rays produced in the <sup>232</sup>Th decay chain are 0.911 MeV (<sup>228</sup>Ac), 0.583 MeV (<sup>208</sup>Tl), and 0.239 MeV (<sup>212</sup>Pb). Uranium is mapped using 1.764 MeV by its daughter element <sup>214</sup>Bi. Other strongest fluxes of gamma rays emitted in the decay chain for <sup>238</sup>U are 1.120, and 0.609 MeV (all from <sup>214</sup>Bi) and 0.352 MeV (<sup>214</sup>Pb).

#### **1.3 GRS Results from Previous Planetary Missions**

Gamma ray spectrometers have been flown in various space missions to explore different planetary bodies such as Moon, Mars, Mercury, and asteroids. Results from gamma spectrometer flown on these missions have been discussed below.

#### **1.3.1 Gamma Ray Experiments on Moon.**

#### • Apollo Mission

Planetary gamma ray spectroscopy was proposed in 1960 by J. R. Arnold to measure the gamma ray radiation coming from moon surface and to find out the

composition of the lunar surface (Arnold et al. 1962). Gamma ray spectrometer is flown by Apollo 15 and 16 with the common objective of geochemical mapping of the lunar surface. About 22% of the lunar surface was mapped (Harrington et. al., 1974) by the combined path flight of Apollo 15 and 16. Elemental maps of Fe, Ti, Mg, K, and Th were produced from the Apollo GRS instrument (Bielefeld et al. 1976, Etchegaray-Ramirez et la. 1983, Harrington et al, 1974, Feldman et al., 1996). From Apollo GRS data, it was inferred that the western mare areas are generally the highest in radioactivity, with the eastern maria being somewhat lower. The highlands are the lowest in activity with a slightly lower level in the far-side highlands. (Apollo 15 Mission Report).

The Apollo 16 results extend and confirm the Apollo 15 finding that all 5° regions within the boundaries of the western maria are higher in radioactivity than any 5° regions outside this area. These results strongly suggest that the western mare regions not flown over are also highly radioactive and that other regions of the Moon are generally less so. The eastern maria show evidence of lower (and variable) radioactivity. The highland regions show low radioactivity, except near the western maria where some lateral mixing has occurred.

#### • Lunar Prospector Mission

The gamma-ray spectrometer in Lunar Prospector (LP) was flown to provide global maps of the elemental composition of the surface layer of the Moon. The main elements mapped are uranium, thorium, potassium, iron, titanium, oxygen, silicon, aluminum, magnesium, and calcium. The LP gamma-ray spectrometer (GRS) consisted of a 7.1 cm diameter by 7.6 cm long cylinder of Bismuth Germanate (BGO) placed within a well-shaped borated-plastic scintillator (BC454) anticoincidence shield (Feldman et al. 1999).

The Lunar Prospector (LP) gamma-ray spectrometer (GRS) has yielded global maps of the elemental composition of the Moon (Lawrence et. Al. 1998). LP measurements have greatly improved the precision of the map and the region covering for more elements. From LP measurements, it has been established that High Alumina (HA) basalts are a more common basalt type and has broadened the surface expanse to at least 180<sup>o</sup> longitudinal (Kramer et. al. 2008). Thorium and potassium maps show that Th and K are concentrated primarily on the nearside within and around the western-most maria and a secondary abundance concentration is located on the farside near Mare Ingenii in the South Pole–Aitken basin (SPA).

Using LP GRS data, Apollo samples, and models of the geophysical structure of lunar impact basins, it has been shown that the Procellarum and Imbrium regions of the Moon are a unique KREEP-rich geochemical province (Jolliff et al, 2000). Furthermore, from these studies, we infer that a substantial portion of the crust in this province is composed of a material similar in composition to Apollo 15 KREEP basalt. The combined study of LP GRS and neutron spectrometer data indicated the enhanced hydrogen deposits near both polar regions. Deposits in the permanently shaded craters near the south pole are consistent with a thick ferroan anorthosite regolith containing an enhancement of 1670  $\pm$  890 ppm hydrogen, which, if in the form of water ice, amounts to 1.5  $\pm$  0.8 % weight fraction of H<sub>2</sub>O (Feldman et a. 2000).

#### • Selene (Kaguya) Mission

The basic scientific goal of the SELENE GRS comprised determination of the elemental composition of the lunar surface, especially for regions unobserved by the Apollo missions. Results from SELENE GRS were important in respect of advancement in research on understanding the origin of the Moon and the evolution of the lunar crust. The GRS observations were also significant for lunar resource exploration, especially regarding the existence of water. Kaguya GRS was a high purity Ge detector, a single n-type crystal, shielded with an active anticoincidence of BGO and plastic scintillators. The Ge detector was of 65 mm diameter, 77 mm length with an active volume of 250 cm<sup>3</sup> relative efficiency 70 %, energy resolution 2.0 keV at 1332 keV (Hasebe et. Al. 1999, 2008). Gamma-ray observation was conducted simultaneously with as many as 14 instruments onboard SELENE, including X-Ray Spectrometer (XRS), Spectrum Profiler, Multiband Imager, Terrain Camera, and Radar Sounder (Sasaki et al., 2003).

Kaguya GRS (KGRS) observed gamma-ray peaks of several major elements including K, Th, U, O, Mg, Al, Si, Ca, Ti, Fe, and Ge. Elemental composition maps for U, Th, and K were derived over the entire lunar surface. From KGRS data, a higher K/Th ratio was observed around Imbrium, with moderate to high Thorium content which could be due to a possible excavation of material different from the KREEP basalts (Forni et. Al. 2010). The Th map from GRS showed that the most rich-area is within Procellarum KREEP Terrain. South-Pole-Aitken (SPA) basin is modestly elevated in Th relative to the rest of the far side. KGRS has provided the global maps of U and K and Th. The KGRS uniquely identified uranium lines from the moon with a superior energy resolution of  $\sim 1\%$ . KGRS provided the global lunar map of uranium distribution for the first time. It was found that uranium abundances vary up to 2 ppm with an average of  $\sim 0.3$  ppm, while the average thorium abundance was  $\sim 1.2$  ppm. From these analyses, significant variation in U/Th ratio was found in the far side of the Moon that had not been reported by previous observations or in lunar materials. From an elemental perspective, the SPA basin region elementally distinct regions as compared to the rest of the moon. The materials within the basin region on average exhibit similar K and Th abundances. Very little change in K and Th counts through time indicate that SPA materials have remained homogeneous relative to K and Th since the basinforming impact (Kim et. al 2012).

Literature survey shows that the SPA is enriched in Fe, FeO, and TiO<sub>2</sub> (Lucey et al., 1995), and Mg (Hiesinger and Head, 2004; Spudis, 2009). From KGRS data it has been found that only minor variations in both Fe and Ti in the SPA region since the formation of the basin, although the observed increase in counts with volcanic resurfacing in the basin. The decrease in the O:Si ratio implies a generally more felsic surface. This could result from resurfacing by ejecta material originating from regions exterior to the basin that source from a shallower depth of the lunar crust. Kaguya GRS data analysis shows that highland terrains are Sienriched relative to lower basins and plains regions, which appear to consist of primarily of mafic rocks (Kim et al. 2013).
#### • Chang'E (Kaguya) Mission

The Chinese Lunar Exploration Program Chang'E, comprised of lunar orbiters, landers, rovers, and in the near future, sample return spacecraft. Chang'E-1 & 2 missions carried gamma ray spectrometers. The Chang'E-1 spacecraft took a gamma-ray spectrometer (CGRS) with the aim of measuring the elemental abundance of major elements, O, Si, Mg, Al, Ca, Ti, Na, and Fe, and of the natural radioactive elements, U, Th, and K upto a depth about 20 cm (Ma et. al. 2008). The energy range of interest was 0.3–9 MeV. Chang'E -1 GRS consists of two large CsI(Tl) crystals as the main and anticoincidence detectors (Zhu et. al., 2010), whereas Chang'E -2 used a LaBr<sub>3</sub> crystal as its main detector and CsI as anticoincidence (Ma et. al. 2013).

Chang'E-1's GRS, IIM (Interference Imaging Spectrometer) and XRS have achieved the abundance of some key elements and distribution of major minerals of the moon (Ouyang et al., 2010, Liu et al. 2010). The IIM data along with GRS was used to detect the chemical and mineralogical compositions of the lunar surface (Zheng et al., 2008; Ling et al., 2011). Ling et al. (2011) developed a new algorithm to map TiO<sub>2</sub> abundance. Highland and mare regional studies suggested that the TiO<sub>2</sub> map has good correlation with Clementine UV-VIS results in corresponding areas. Chang'E-1 GRS results show that high-radioactive composition is concentrated in the Procellarum KREEP Terrane (PKT) on the nearside. GRS measurements illustrate that SPA basin has moderate concentration and the highlands have a low concentration of radioactive elements. The inverse correlation in high lands of PKT and much of SPA basin suggests radioactive-rich materials increasing in abundance at lower depths (Zhu et. al., 2010).

The K distribution from CE-2 GRS is in general agreement with LP, Kaguya, and Chang'E-1 measurements. The global average abundance of K is  $620 \pm 615$  ppm with the upper limit of 3240 ppm, which is lower than the LP value of 755  $\pm$  740 ppm with the upper limit of 4360 ppm (Zhu et al 2013). Chang'E-2 Gamma Ray Spectrometer was used to derive the lunar Fe map, which shows that majority of iron-rich regions match well with mare basalt deposits. Oceanus Procellarum and central Mare Imbrium dominate the high values (FeO-23 wt.%) of iron

abundance and the richest FeO (30.2 wt.%) region locates in the young mare basaltic unit south of Aristarchus crater. South Pole-Aitken basin is relatively enriched in iron (maximum of 12.7 wt.%) compared to the highlands poor of iron (FeO 8 wt.%). GRS based global Al<sub>2</sub>O<sub>3</sub> map shows the diverse surface of the Moon: significant maria-highlands dichotomy; heterogeneity among mare units or highlands. On the nearside, mare could be separated from highlands with the contour line of 16 wt.% Al<sub>2</sub>O<sub>3</sub>, while the threshold value rises to 21 wt.% on the farside.

#### **1.3.2 Gamma Ray Experiments on Mars.**

Metzger and Arnold have discussed the use of gamma ray spectroscopy at Mars (Metzger and Arnold, 1970). On board gamma ray experiments (using CsI(Tl) from Phobos 2 provided few quantitative results for elemental abundances of Martian surface, (Surkov et al. 1989 & Trombka et al. 1992). The Mars Odyssey GRS experiment is, therefore, the first elemental chemical mapping instrument to successfully orbit Mars.

The primary scientific objective of Mars Odyssey GRS was global mapping of major and minor elements on the Martian surface (Boynton et al 1992, 2004). A neutron spectrometer along with GRS provided information about the abundance of hydrogen in shallow Martian subsurface (upto  $\sim 60$  cm). The abundances of refractory incompatible elements like Th and U was considered to provide particularly useful information about the degree of fractionation of the Martian crust with respect to the mantle. The abundances of radioactive Th, K and U in the Martian crust are important as constraints on the magnitude of radiogenic heating sources present, and K/U and K/Th ratios may be used to infer the abundances of K and other moderately volatile elements on the planet.

Mars Odyssey orbiter GRS provided elemental abundance maps for H, Si, Fe, Cl, K, and Th in the low-latitude and mid-latitude regions of Mars, (Boynton et al., 2004, 2007). The elemental map of Si shows a range from 18.5% to 21.5%. Iron (Fe) map shows an enrichment of Fe in the northern lowlands. Mars GRS measurements show a variation of Fe concertation from 14% to 19% in the

lowlands while it is 11% to 14% Fe in the highlands (Boynton et. al. 2007). For the first time, Mars Odyssey GRS measurements provided the distribution of Cl at the equator and mid-latitudes of Mars (Boynton et. al. 2007 & Keller et.al. 2006). The map of Cl shows significant variation over the mid latitudes, and a significant enrichment on the west of the Tharsis Montes and northeast of Apollinaris Patera is observed. Cl is significantly enriched at the surface relative to the Martian meteorites.

Many evidences have previously suggested aqueous activity on Mars and geochemical studies show that K/Th is affected by rock-water interactions. Mars Odyssey GRS observation shows that K/Th does not change significantly on the surface of Mars; K/Th ranges generally between 4000 and 7000. As compared to Earth, bulk Mars is enriched in moderately volatile elements but has a much lower K/Th. GRS data shows that K and Th abundances decrease most significantly from the Hesperian to the Amazonian age categories, with an approximate 9% decrease in K (from  $3340 \pm 20$  to  $3070 \pm 20$  ppm) and 7% decrease in Th (from  $0.64 \pm 0.03$  to  $0.61 \pm 0.03$  ppm) (Taylor et al. 2006a, 2006b).

#### **1.3.3 Gamma Ray Experiments On Mercury**

MESSENGER Gamma-Ray Spectrometer (GRS) is designed to identify elements within tens of centimetres of the surface by remotely detecting their characteristic gamma-ray emissions in the 0.1- 10 MeV energy range and by characterizing the flux of escaping thermal, epithermal, and fast neutrons (Goldsten et al., 2007 & Evans et.al. 2012). The MESSENGER GRS measurements have provided the abundances of several major elements (Al, Ca, S, Fe, and Na) on Mercury. GRS data suggests that Mercury's regolith is, on average, vertically homogeneous to a depth of tens of centimeters. The GRS and XRS elemental results suggest that Mercury's surface is dominated by Mg-rich silicates. The Na abundance is estimated to be  $\sim$ 3% which suggests that plagioclase feldspar on Mercury's surface is relatively rich in albite (Evans et. al. 2012). Mercury's surface mineralogy is dominated by high-Mg mafic (mostly enstatite), and plagioclase feldspar (calcic and sodic) phases, with smaller amounts of sulfide minerals (e.g., old hamite). The smooth plains within the Caloris basin are similar in composition to the northern plains (Weider et. al. 2012).

MESSENGER GRS measurements shows an average K/Th ratio on the surface of Mercury (north of  $\sim 20^{\circ}$ S) and this value is comparable with that of other terrestrial planets. K/Th of Mercury is an order of magnitude higher than lunar values, whereas the absolute abundances of K and Th are 3 – 4 times lower than on the surface of Mars. GRS measurements of the abundances of the radioactive elements Th, K, and U indicate that Mercury's interior heat production may have been four times higher  $\sim$ 4 Ga than at present (Peplowski et al., 2011, 2012). A new technique (estimating the altitude dependence GRS signal to reduce the large background) has been applied to analyze Al data measured by the MESSENGER GRS to determine the abundance of Al on the surface of Mercury. The estimated result from MESSENGER GRS confirms the finding of low abundance of Al by X-Ray spectrometer. The low abundance of Al rules out a global, lunar-like feldspar-rich crust and is consistent with previously suggested analogs for surface material on Mercury, including terrestrial komatiites, low-iron basalts, partial melts of CB chondrites, and partial melts of enstatite chondrites (Peplowski et. al. 2012).

For the first time, the abundance of chlorine on the surface of Mercury has been determined by MESSENGER GRS measurements. The results show that Cl abundance is ~3 times greater at high northern latitudes than in mid and equatorial latitude bands. GRS results also provided the evidence of volatile elements (e.g., S, K, Na, and Cl) are not depleted on the surface of Mercury, (Evans et. al. 2015), and instead are present at abundances similar to those on other terrestrial planetary bodies. Mercury has a chondritic Cl/K ratio, indicating that it has not experienced loss of volatile elements through collisional erosion (Evans et. al. 2015).

#### 1.3.4 Gamma Ray Experiments on Dawn and Ceres (Asteroids)

The Dawn mission aimed at exploration of two of the largest main belt asteroids, 1 Ceres and 4 Vesta. Dawn Gamma Ray and Neutron Spectrometer has measured the averaged Fe/O and Fe/Si abundance ratios of Vesta's regolith on a global scale and a global distribution of H (using neutron spectrometer). These measurements revealed that diogenite-like material is exposed within the Rheasilvia impact basin (Prettyman et al. 2012). These results show that the elemental composition of the surface of Vesta is consistent with the howardite, eucrite, and diogenite (HED) meteorites which is consistent with earlier terrestrial-based spectral studies measurements (McCord et al. 1970). GRaND has provided the globally averaged concentrations of radioelements K and Th within Vesta's regolith for the data set acquired by Dawn during 5 months of low-altitude operations (Prettyman et al. 2015). The concentration of K and Th measured by GRaND within Vesta's global regolith are consistent with eucrite-rich howardite and are distinct from most achondrites, all chondrites, and Mars meteorites. The K/Th ratio of Vesta (900  $\pm$  400) is similar to the average ratio for howardite (approximately 1200).

From Dawn GRaND, the estimation of Fe abundances for Ceres was carried out by measuring 7.6 MeV gamma ray produced by neutron capture. Whereas at equator, the average Fe was estimated to 16  $\pm$ 1 % (Prettyman et. al. 2016 & Yamashita et. al. 2013), a longitudinal variation was observed for Hydrogen abundances suggesting that the processes responsible for the presence of Hydrogen on Ceres surface have acted on a global scale (Prettyman et. al. 2016 & Yamashita et. al. 2013). GRaND measurements show that Ceres contains a greater amount of C as compared to carbonaceous chondrites, while the K contain is similar to that of CM and CI chondrites (Yamashita et. al. 2013). At ~300 km spatial scale GRaND measurements show that basaltic eucrite occurs only ~3% of the Vesta surface, cumulate & ortho-pyroxenenitic diogenite are not detected (Beck et.al. 2017).

#### **1.4 Motivation of the Present Thesis**

Gamma ray spectrometer has been flown in various space mission to explore different planetary bodies like Moon, Mars, Mercury, and asteroid. However, as the 7.6 MeV gamma lines from iron were not fully resolved from a 7.72 MeV aluminum line, one cannot rule out the contribution of the Al line to the observed signal, particularly in view of the fact that Lunar Prospector iron map contained regions with anomalously high Fe abundances. Iron abundance maps over the entire moon have been previously obtained using Clementine Spectral Reflectance (mineralogy) data and from the Lunar Prospector gamma spectrometer data. However, the Prospector iron map contains regions with anomalously high counting rates, possibly due to a significant contribution from an unresolved interference from an aluminum line.

Quantifying secular variations in the chemical composition of the Martian crust provides unique insights into the processes that have guided the evolution of the Martian crust-mantle system. Gamma Ray Spectrometer (GRS) data can be used to understand the evolution of planets. For example, Mars Odyssey GRS provided an average abundance of K, Th, Fe, Cl, H, and Si for the major Martian geologic epochs (Noachian, Hesperian, and Amazonian). Average GRS-determined K and Th abundances generally decrease by 9% and 7%, respectively, between the Hesperian and the Amazonian, possibly implying evolving magma chemistry throughout major resurfacing events (although the effects of surficial alteration processes cannot be entirely discounted). GRS-determined Fe and Cl averages increase by 12% and 19%, respectively, with younger apparent relative surface age, suggesting the possible mobilization and transport of these elements through aqueous processes (although an igneous origin for the variation in Fe also cannot be excluded). While H abundance does vary with surface age, the relationship is likely not governed by geologic processes. No statistically reliable apparent surface age relation was found for Si.

The Mars Odyssey GRS instrument (Boynton et al. 2007) provided concentration maps for the elements H, Si, Cl, K, Fe and Th for low and midlatitudes (±45°) for the first time. However, the Fe concentrations obtained by GRS were 20% higher than measurements based on Mars landing missions (Viking, Pathfinder, Spirit, and Opportunity). Furthermore, no maps have been reported for Ca, Al, S, U, Na, and Mn from Odyssey GRS data. An interesting scientific implication from Odyssey GRS data is that there exists no evidence for a globally distributed dust of homogeneous composition on Mars with a thickness of the order of tens of centimeters.

In light of the above, the present study focused on development of the gamma ray spectrometer for future planetary missions, for mapping of naturally radioactive elements (Th, U, K), and other major and minor elements including H, O, C, Fe, Mg, K, Ti, Ca, Al, Si, S on the entire Martian/Moon surface.

#### **1.5 Research Objectives of the Present Study**

The research objectives of the present study are the following:

- Development of front-end-electronics and processing electronics subsystems for a LaBr<sub>3</sub>:Ce gamma ray spectrometer
- Testing and Calibration of the LaBr<sub>3</sub>:Ce gamma ray spectrometer
- Simulation of LaBr<sub>3</sub>:Ce gamma ray spectrometer response to photons and high energy protons using Geant4 simulation toolkit
- Development of front-end-electronics and processing electronics subsystems for a CeBr<sub>3</sub> gamma ray spectrometer
- Testing and Calibration of the CeBr<sub>3</sub> gamma ray spectrometer
- Application of HPGe gamma spectrometer for estimation of annual doserate for terrestrial samples

#### **1.6 Scope of the Present Thesis**

When a gamma ray photon interacts with a LaBr<sub>3</sub>:Ce crystal, the energy of the gamma photon is converted into a light pulse. The scintillation light has a maximum emission at 380 nm, and is converted to electrons in a PMT, which also amplifies the electron current by 5-6 orders of magnitude. The charge pulse is collected by a charge integrating preamplifier which generates a voltage pulse whose height is proportional to the energy deposited in the crystal. The pulse is then shaped and amplified further by a linear amplifier from a few hundred millivolts to a few volts for A/D conversion. Thus digital information of the photon energy is interfaced with S/C using FPGA based digital control system.

Three types of detector systems are suitable for the energy range 0.1 to 10 MeV:

- (i) 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, typically ~10-12% at 662 keV (<sup>137</sup>Cs) gamma ray line, and are not considered for the present experiment.
- (ii) High Purity Germanium (HPGe) detectors have been used in the 2001 Mars Odyssey Mission and in the recent Selene (Kaguya) moon 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. The FWHM (energy resolution) for this detector was 4.1 keV (~0.3%) at the 1332 keV line from <sup>60</sup>Co. However, the HPGe detectors need to be cooled to liquid nitrogen temperatures (77 K) in order to achieve high energy resolution levels, implying the use of a Stirling cycle cooler or a passive cooling system. Such cooling devices are heavy and require a long time for development. Sterling refrigerators require about 50 W of power. Thus, limitations on weight and power make this detector unsuitable for this moon mission.
- (iii) The newly developed cerium doped lanthanum bromide (LaBr<sub>3</sub>: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 LaBr<sub>3</sub>:Ce detector is superior to NaI(Tl). The energy resolution of this detector is 2.8% at 662 keV (<sup>137</sup>Cs) and ~1.6% at 2615 keV (<sup>208</sup>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.

Based on weight, power, and operating temperature considerations for spacecraft payloads, a LaBr<sub>3</sub>:Ce gamma detector would be the best choice for the gamma spectrometer and is our choice for the proposed gamma ray spectrometer. However, due to large intrinsic activity, LaBr<sub>3</sub>:Ce gamma spectrometers were not very useful in measurement of U, Th and K concentrations. Thus, a CeBr<sub>3</sub> gamma spectrometer has been developed in-house to estimate the concentration of K, U and Th for rock/soil samples.

### Chapter – 2

# Gamma Ray Spectrometry Principles and Detection Techniques.

#### 2.1 Principle

#### 2.1.1 Radioactive Decay

Radiation produced in atomic or nuclear processes can be classified as charged particulate radiation or uncharged radiation. The charged particulate radiation includes the fast electrons ( $\beta$  particles emitted in nuclear decay) and heavy charged particles ( $\alpha$ , protons, and fission products etc.), while electromagnetic radiation (X-rays,  $\gamma$ -rays) and neutrons comprise uncharged radiation (Knoll, 1999). These nuclear decays provide the information of nuclear structure through the measurement of the state's excitation energy and decay rate. The gamma-rays are basically uncharged photons produced from nuclear reactions.

#### Law of Radioactive Decay

Radionuclides are unstable and decay of one or more of the decay modes; alpha, beta minus, beta plus or electron capture. The amount of a radionuclide in a sample is expressed in Becquerel numerically equal to the rate of disintegration the number of disintegrations per second. This is known as the activity of the sample. The rate of decay is directly proportional to the number of atoms of the radionuclide present in the source, i.e. the activity, A, is directly proportional to the number of atoms, N, of nuclide present:

$$A = -\frac{dN}{dt} = \lambda N \tag{1}$$

Integrating the above equation leads to the exponential law of radioactive decay and can be written as

$$N(t) = N_0 e^{-\lambda t}$$

where N<sub>0</sub> is the number of atoms at time t = 0 and  $\lambda$  proportionality constant.

The proportionality constant,  $\lambda$ , is called the decay constant (the probability per second that the emission takes place) and has the units of reciprocal time (s<sup>-1</sup>). The reciprocal of the decay constant is the mean lifetime,  $\tau$ , of the radionuclide, the average time which an atom can be expected to exist before its nucleus decays:

$$\tau = \frac{1}{\lambda}$$

This time represents a decay of the source by a factor of e (i.e. 2.718). It is more convenient and meaningful to refer to the half-life,  $t_{1/2}$ , of the radionuclide – the time during which the activity decreases to half its original value:

$$t_{1/2} = \frac{\ln 2}{\lambda} = \frac{0.693}{\lambda}$$

In practice, it is more useful to replace the number of atoms by activity, bearing in mind that activity is proportional to the number of atoms:

$$A(t) = A_0 e^{-\lambda t} \tag{3}$$

The activity is expressed as the unit of Curie (Ci), which is defined as  $3.7 \times 10^{10}$  disintegrations / second (dps), which is defined as estimation of activity of 1 gram of pure <sup>226</sup>Ra. The curie SI equivalent, is expressed in terms of the Becquerel (Bq), thus:

1Bq = 1 dps

#### 2.2.2 Gamma Decay

The decay of an excited nucleus loses its energy by the transition from a higher energy state to a lower energy state. The released energy appears in the form of a gamma ray photon. The other mode of gamma decay is the internal conversion process in which the excited nucleus de-excites by ejecting an electron from one of the orbits. The lifetimes of these processes are usually very short compared with  $\alpha$ - or  $\beta$ -decay lifetimes and are typically, less than 10<sup>-9</sup> s.

#### 2.2 Interaction of Gamma Rays with matter

Gamma rays cannot ionize the absorbed material directly but interacts with the absorber in different ways depending on its energy. There are three main principal processes by which gamma- ray photons interact with the matter: *photoelectric absorption, compton scattering* and *pair production*.

#### 2.2.1 Photoelectric Absorption

Photoelectric absorption is the process in which the incident gamma ray photon is completely absorbed when it interacts with the atoms of the absorbing material. This energy is transferred an orbital electron by injecting an energetic photoelectron from the atom. The energy of the photoelectron can be expressed as.

$$E_{e^{-}} = h \nu - E_b \tag{4}$$

Where  $E_b$  is the binding energy of the electron.

The photoelectron is generally emitted from the K-Shell for which binding energy is in few keV for low Z-element and tens of keV for high-Z element. Due to the process of photoelectric emission a vacancy is created which is filled by either capturing nearby free electrons or rearrangement of an electron from other orbit shells of the atom. In this rearrangement process, the binding energy is released in the form of a characteristic X-ray. Figure 2.1 shows the schematic of photoelectric effect.



#### Figure 2.1: Schematic of representation of photoelectric absorption

The photoelectric process is predominant for low energy gamma ray and is enhanced for absorber materials of high atomic number. The probability of photoelectric absorption per atom can be expressed as

$$\tau \cong \propto Z^n E_{\gamma}^{-3.5}$$

-----(5)

Where, Z is atomic number and n ranges between 4 and 5.

#### 2.2.2 Compton Scattering

Compton scattering is the process in which the gamma ray photon interacts with a single free electron and abruptly changes its direction transferring a portion of its original energy to the electron from which it scattered. The energetic electron is called as Compton electron or recoil electron while the deflected photon with rest of energy is called scattered photon (Knoll, 1999). Figure 2.2 shows the Compton scattering process.

The recoil electron turn out to be a free electron with kinetic energy equal to the difference of the energy lost by the  $\gamma$  ray and the binding energy of the electron. Because the binding energy of the electron is very small compared to the  $\gamma$ -ray energy, the kinetic energy of the electron is very nearly equal to the energy lost by the gamma ray:

$$E_e = E_\gamma - E' \tag{6}$$

Where, Ee is the energy of the recoil electron,

 $E_{\gamma}$  is the energy of the incident gamma ray, E'is the energy of the scattered photon, and E' can be expressed as

$$E' = \frac{E_{\gamma}}{1 + \frac{E_{\gamma}}{m_0 c^2} (1 - \cos\theta)} \tag{7}$$

Where,  $m_0c^2$  is rest mass energy for an electron (511 keV)

 $\theta \rightarrow$  Angle between the incident gamma ray and the scattered photon.



Figure 2.2: Schematic of representation of Compton scattering process.

The probability of Compton scattering per atom of the absorber depends on the number electrons available as scattering targets. Therefore, this increases linearly with Z (atomic number).

#### **2.2.3 Pair Production**

When the energy of the gamma ray > 1.022 MeV, pair production may occur as an alternative to the Compton process. The probability of this interaction is very low until the  $\gamma$ -ray energy exceeds several MeV an therefore this interaction is mostly restricted to high energy gamma rays. In this interaction the energetic photon comes close to a nucleus and transformation of energy into mass occurs because of the electric field of the nucleus. With this, the photon is converted into an electron and a positron with the same mass.

The importance of all the three process described earlier for different absorber over a range of gamma energy has been represented in Figure 2.3.



Figure 2.3: Relative importance of tree types of gamma ray interaction.

#### 2.2.4 Total Absorption

The above mentioned type of interaction depends on the photon energy and the material used. These interactions can be characterized by a fixed probability of occurrence per unit length. The sum of these probabilities can be expressed as

$$\mu = \mu_{Photo} + \mu_{Comp} + \mu_{Pair} \tag{8}$$

and is called the mass attenuation coefficient.

The number of incident photons can be expressed as

$$I = I_0 e^{-\mu\rho x} \tag{9}$$

Where,  $\rho$  and x are the density of the medium and path length respectively.

#### 2.3 Radiation Detector

The principle of detection of radiation is based on its interaction and the energy deposited in the material of which the detector is made. When neutral radiation particle like X-rays,  $\gamma$ -rays interacts with the detector material, they produce secondary electrons and energy is deposited in the detector. The selection of detector depends upon the type of radiation to be detected. Detection of gamma ray requires the following characteristics:

- High efficiency of detection
- High energy resolution.
- Linearity
- Good mechanical and electrical stability.

#### 2.3.1 Types of Radiation Detector

The radiation detectors are operated on the basis of the way the photons interact with the matter which constitutes the detector, in manner that all the part of energy possessed by the radiation is transferred to the electrons and is collected as an electrical signal. There are different types of detector

- I. Gas Filled Counters
- II. Semi-conductor detector
- III. Scintillation detector

Out of these three detectors Semi-conductor detector and scintillation detector are mostly used for the detection of gamma ray.

#### (I) Gas Filled Counters

The primary modes of interaction involve ionization and excitation of gas molecules along the passage of radiation, but generally most gas filled detectors are based on direct ionization created by the radiation photons. The ion pair produced by ionization move under the influence of electric field until recombination takes place or lose their energy through a collision as shown in Figure 2.4. This motion induces a current which is measured through an electronic device.



Figure 2.4: Schematic of gas filled detector.

#### (II) Semi-Conductor Detector:

The operating principle of semi-conductor detector is same as gas detector. In semiconductor the charge carrier are electrons and holes instead of electrons and ions. The radiation photon incident on the semiconductor junction produces an electron-hole pair which move away under the influence of applied electric field as shown in Figure 2.5. Pre-amplifier collected these charges in the form of pulses.



Figure 2.5: Schematic semiconductor detector.

#### (III) Scintillation detector

The detection of ionized radiation by scintillation material is one of the most popular techniques. Scintillation material converts the energy of the ionized radiation into a flash of light. Out of these three detectors semi-conductor detectors (mostly High Purity Germanium (HPGe) and scintillation detectors (Mostly NaI(Tl), BGO, CsI(Tl)) are commonly used for detection of gamma ray. The advantage of semiconductor detector is that they provide very good energy resolution provided the detector is operated at liquid nitrogen cooling temperature, in contrast to scintillation detectors which can be operated in room temperature. This thesis work is focused on development of gamma ray spectrometer using new scintillation detectors. The detail principle and mechanism of scintillation process has been discussed.

#### 2.4 Characteristics of Gamma Ray Spectrometer

In gamma-ray spectroscopy applications, the magnitude of the output pulses produced by the detector are proportional to the energy deposited by the incident photons in the detecting medium. The pulse-height spectrum is a direct depiction of the energy spectrum of the  $\gamma$ -ray interactions and provide spectroscopic information.

#### 2.4.1 Detector Response

Irrespective of the type of detector used, there are many common features in the measured spectrum. Figure 2.6 shows a gamma spectrum from a monoenergetic gamma-ray source of energy E. The gamma ray photon detected by the detector do not usually come from free nuclei. They are emitted in material media, some of them endure scattering before they emerge from the radioactive source. Due to the scattering the affected photons lose some energy and therefore the energy spectrum of photons emitted from the sample is slightly broadened into energies below peak energy. When the gamma ray interacts in the detection medium, it transfers part or all of its energy to an atomic electron and a free the electron is emitted from its atomic bond. This free electron transfers its kinetic energy, in a series of collisions, to other atomic electrons in the detector medium.

Photoelectric interaction transfers all of the incident photon's energy to a photoelectron which then subsequently causes multiple ionizations until its energy is depleted. Therefore, the amount of charge produced from this event is proportional to the actual photon energy. In Compton scattering interaction only a part of incident photon's energy is transferred the to an ionized electron and that electron subsequently cause the ionization until its energy is exhausted. The amount of charge produced from this event is proportional to the partial energy formerly lost by the incident photon but this does not convey useful evidence about the actual photon energy. In Compton scattering event, the maximum energy can be deposited in the detection medium when the photon is scattered by 180°. Hence, the Compton-generated pulses are distributed below this maximum energy and constitute a source of "background". The full-energy peak is considerably broadened by the statistical variability in the number of electron-ion pairs produced by the photoelectron. This effect is the primary contributor to the width of the full-energy peak and is therefore the dominant factor in the detector energy resolution.



## Figure 2.6: Representation of the gamma detector spectrum from a mono-energetic gamma source.

- **Photopeak**. This peak characterizes the pulses that arise from the fullenergy, photoelectric interactions in the detection medium. The width of the peak determines the statistical instabilities in the charge produced from the interactions. The centroid of the peak represents the photon energy E<sub>0</sub>.
- **Compton plateau.** Compton pulses, distributed smoothly up to a maximum energy E<sub>c</sub>, originate from interactions including only partial photon energy loss in the detecting medium. Compton events are the primary source of background counts under the full-energy peaks.
- The Compton Edge. This is the region of the spectrum which indicates that the incident photon has lost its maximum energy through Compton scattering. It is a broad asymmetric peak corresponding to the maximum energy (E<sub>c</sub>) that a gamma ray photon of energy E can transfer to a free electron in a single scattering event.
- **Compton valley:** For a mono-energetic source, pulses in this region arise from either multiple Compton scattering events or from full-energy interactions by photons that have undergone small-angle scattering before entering the detector.

- Backscatter Peak. This peak is instigated by gamma rays that have first interacted by Compton scattering in one of the materials surrounding the detector. Gamma rays scattered by more than 110°-120° will emerge with nearly identical energies. This peak is mostly located in the vicinity of 200
   250 keV. Therefore, a mono-energetic source will give rise to many scattered gamma rays whose energies are near this minimum value.
- Low-Energy Rise. This feature of the spectrum, very near the "zero-pulseheight-amplitude" region, arises typically from low-amplitude electronic noise in the detection system that is processed like low-amplitude detector pulses. This noise tends to be at rather high frequency and so appears as a high-count-rate phenomenon.

#### 2.4.2 Energy Resolution

The energy resolution is the capability of the detector to isolate radiation of different energy. As shown in Figure 2.7, the energy resolution can be measured by the full width at half maximum (FWHM) which represents the peak width. Therefore, the energy resolution R can be expressed as a percentage and is determined by the following equation:

$$R = \frac{\Delta E}{E} = \frac{FWHM}{H_0} \times 100 \%$$
 ------(10)

where Ho is the channel number corresponding to position of the peak centroid.



Figure 2.7: Definition of detector energy resolution.

For a Gaussian shape peak, the FWHM can be expressed as 2.35 times the standard deviation. The statistical broadening of the energy resolution varies with gamma ray energy. The FWHM of the peak is proportional to the square root of the gamma ray photon energy. The gamma ray energy is proportional to the average pulse height produced. Therefore, from the definition the energy resolution can be express as the following relation:

$$R = \frac{k}{\sqrt{E}} \tag{11}$$

Where, k is a constant of proportionality.

The above equation can be represented as,

$$\ln R = \ln k - \frac{1}{2} \ln E$$
 (12)

The plot of this equation is a straight line with a slope of -1/2.

#### 2.4.3 Detection Efficiency

Gamma rays (photons) may travel inside the detector without any interaction which means that the detector is less than 100% efficient. Therefore, it is necessary to relate the number of pulses counted to the number of gamma ray incident on the detector. The detector efficiency depends upon

- Type and energy of radiation
- Density, size of detector material and distance between the source and the detector
- Electronics

In general, there are three main types of efficiency namely, (i) absolute efficiency (ii) intrinsic efficiency and (iii) photopeak efficiency.

#### (i) Absolute Efficiency (Et)

Absolute total efficiency is the number of counts recorded by the detector per unit time  $C_t$  over the number of gamma ray emitted by the source per unit time Ny and can be calculated by using the following equation:

where  $N\gamma$  can be derived by:

where  $D_s$  is the current activity of the source and I $\gamma$  ( $E_{\gamma}$ ) is the fractional number of gamma rays emitted per disintegration.

#### (ii) Intrinsic Efficiency (Ei)

The intrinsic total efficiency can be defined as the number of pulses produced by the detector to the total number of gamma ray incident on the detector. This efficiency can be calculated by using the following equation:

$$\varepsilon_i = \frac{4\pi}{\Omega} \varepsilon_t \ x \ 100 \ \% \tag{15}$$

where  $\Omega$  is the solid angle subtended by the detector at the point source.

#### (iii) Photopeak Efficiency (*E*<sub>P</sub>)

The photopeak efficiency is the probability that a gamma ray at a specific energy which is incident on the detector will be recorded in the photopeak. This efficiency is expressed as:

where  $C_P$  is the net number of counts under the full energy peak per unit time.

#### 2.5 Analysis of Pulse Height Spectra

The pulse amplitude is commonly represented through differential pulse height distribution as shown in Figure 2.8. The peak height provides the energy information while, the total counts in the peak area can provide the concentration information.



Figure 2.8: Pulse height distribution of gamma ray spectrum.

The net counts in the peak area N can be calculated as,

$$N = C - B$$
  
=  $\sum_{i=l}^{i=k} C_i - \sum_{i=l}^{i=k} B_i$  .....(17)

Where C is the total count between the region l to k and B is the background under the same region of interest.

The background can be calculated by choosing the base line from the peak and integrating the counts under the line gives the background counts. Otherwise also the background can be calculated as by summing the total counts upto a fixed number of channel after and before the region of interest. In this case background can be expressed as

#### Gamma ray counting statistics

The gamma ray measurements each event are independent of each other and hence these applications normally meet the conditions that define the Poisson Distribution. The events are uniformly and randomly distributed over the sampling intervals. If the events are counted over a finite time period, t, the Poisson Distribution, P(N), describes the probability of recording N counts in a single measurement of duration, t.

$$P(x) = \frac{(\mu)^{-x} e^{-\mu}}{x!}$$
(19)

The mean of the event can be expressed as  $\langle x \rangle = N = \mu$  with standard deviation can be found as  $\sigma^2 = N = \mu$ .

#### 2.6 Current Development in Scintillation Crystals

In the 1940's, sodium iodide, essentially the first scintillator was produced for radiation detection and still remain the commonly used detector for the detection of gamma rays. This is because sodium iodide, a common salt, is relatively inexpensive, environmentally robust, has favorable fabrication and can be grown in a cubic meter sizes. However, it is lacking in some desirable features, in particular the speed of its signal, its energy resolution and its effective atomic number. As a result, a number of new scintillators are emerging that improve in one or more of these categories [Wehe 2006].

In search of good scintillation crystal, over the past few years a lot of research have been focused on the development of new scintillation crystals and a number of interesting materials have emerged as scintillators that utilize Ce-like atoms as the primary scintillation source. These have included YAP(Yb<sup>3+</sup>) (i.e., YAlO<sub>3</sub>), YAG, and LuAP, all of which have light yields which are only about half that of NaI, but are significantly faster (Moses, 2002). The biggest breakthrough is the development of rare earth halide crystals, RX<sub>3</sub>, where R is La, Lu, Ce etc. and X is for I, Br, Cl. Among these halide crystal Ce<sup>+3</sup> activated LaCl<sub>3</sub> and LaBr<sub>3</sub> scintillators, are the best one (Shah et al. 2003). These two materials not only provide much

faster signals ( $\sim$ 30 ns), but also high light output and much better energy resolution. CeBr<sub>3</sub> another rare halide crystal which also provide faster signal ( $\sim$ 17 ns), but the light output and the energy resolution are not better than LaBr<sub>3</sub>:Ce.

For this thesis work LaBr<sub>3</sub>:Ce and CeBr<sub>3</sub> have been used to develop the gamma ray spectrometer and will be discussed in Chapters 3 and 4. The performance of these detector has been simulated using a Geant4 simulation tool kit.

#### 2.7 Simulation of Performance of Scintillation Detector

#### 2.7.1 Geant4 Simulation Tool Kit

GEANT4 stands for "Geometry and Tracking", based on object oriented methodology and C++ language. Geant4 is a toolkit developed by engineers and scientists at CERN and other institutions worldwide. Geant4 is developed to meet the large-scale, accurate, and comprehensive simulation of the particle detectors used in modern particle and nuclear physics (S. Agostinelli et.al 2003). Geant4 is based on set of physical models to handle the interaction between the incident particle and the matter trough a wide energy range from the eV to TeV scale. Users can construct stand-alone applications or applications built upon another objectoriented framework. In nuclear physics, we are very much familiar with interaction of radiation (e.g. alpha, beta, electron, positron, x-rays, gamma-rays etc.), with matter. GEANT4 provides an efficient way to visualize these processes.

Geant4 is driven by the software needs of modern experiments. A typical software system contains components event generator, detector simulation, reconstruction and analysis that can be used separately or in combinations. The toolkit has a hierarchical structure of domains that are linked together when the program is compiled with a GNU make file. Several domains that are necessary to construct a physics case include the following:

- The geometry and material of the detector used.
- Particle interaction within the detector medium or other matter
- Tracking of the particle. i.e. how many steps to move the particle.

- Digitization and hit management
- The hit pattern, event and track management
- visualization and visualization framework
- user interface.

To understand the overall performance of the newly developed scintillation detector, the simulation work has been carried out to determine the absolute efficiencies of the detector for close source-to-detector geometry. Experimental efficiency calibration is a difficult and time-consuming procedure for each specific geometry. Whereas, Monte Carlo methods are based on determination of full energy peak (f.e.p.) efficiency by simulating all relevant physical processes taking place along the path of a photon emitted by the source (S. Hurtado 2004). The history of each individual primary particle consists of its emission by the source, interaction with the detector and surrounding materials, production as well as transport of secondary particles, and track until the photon escapes or undergoes a photoelectric interaction in the crystal, depositing all of its energy.

#### 2.7.2 Detector Geometry

In this simulation study, three scintillation detectors LaBr<sub>3</sub>:Ce, CeBr<sub>3</sub> and NaI(Tl) have been taken into consideration. The simulation also has been carried out for by changing the volume of the detector. In this study, Geant4 simulation has been carried out for three different detector sizes, 1" x 1", 2" x 2" and 3" x 3". The simulation has been carried out by considering the detector geometry as cylindrical shape. In Figure 2.9 shows a geometry of a 3" diameter x 3" length LaBr<sub>3</sub>:Ce covered with a 0.5 mm thick Al casing constructed the simulation. Figure 2.10 shows the simulation of interaction of beam with the detector.



Figure 2.9: Construction of 3" x 3" detection with Geant4 simulation with 0.5 mm Al casing.



Figure 2.10: Interaction of beam with LaBr<sub>3</sub>:Ce Detector.

#### 2.7.3 Efficiency of LaBr<sub>3</sub>:Ce, CeBr<sub>3</sub>, and NaI(Tl) Detector

The efficiency corresponds to the number of particles detected compared to the number of particles emitted. The absolute efficiency and peak efficiency are defined as

$$\varepsilon_{abs} = \frac{Total \, number \, of \, gamma \, detected}{Total \, number \, of \, gamma \, emitted \, from \, source} \times 100 \,\%$$

.....(20)

The photopeak detector efficiency can calculated

$$\varepsilon_{\text{peak}} = \frac{\text{no of gamma detected under the photopeak}}{\text{no of gammas emitted by source}} \times 100\%$$

.....(21)

Several gamma energies (10 keV to 10 MeV) have been implanted on the detector surface and the spectra has been collected for each energy. The spectra have been generated by implanting 10<sup>2</sup>, 10<sup>3</sup>, 10<sup>4</sup>, 10<sup>5</sup> and 10<sup>6</sup> particles on the surface. It has been found that for all the detector when the count rate has been increased the efficiency also increases hence, the results obtained with 10<sup>6</sup> particles has been discussed in this section. The photo-peak efficiency of the detector has been calculated for each energy. Figure 2.11 shows the plots obtained for energy vs efficiency for different size of LaBr<sub>3</sub>:Ce. The same has been repeated for CeBr<sub>3</sub> and NaI(Tl) detectors, and the energy vs efficiency plots are shown in Figures 2.12 and 2.13 respectively. The efficiency calculated from the simulated results of LaBr<sub>3</sub>:Ce, CeBr<sub>3</sub> and NaI(Tl) for 3″ x 3″ detector size has been compared. The efficiency is calculated over an energy range from o.1 to10 MeV and is shown in Figure 2.14 and efficiency of few energies of all three detectors shown in Table 2.1.

Energy in keV	LaBr <sub>3</sub> :Ce	CeBr <sub>3</sub>	NaI(Tl)
100	0.97	0.97	1.00
300	0.94	0.94	0.82
500	0.81	0.81	0.56
1000	0.57	0.58	0.32
1500	0.45	0.46	0.23
3000	0.29	0.30	0.13
7000	0.17	0.18	0.06
8000	0.16	0.16	0.05

 Table 2.1. Comparison of efficiency for 3" x 3" scintillation detectors.



Figure 2.11: Photo-peak efficiency simulated for different size of LaBr<sub>3</sub>:Ce detector.



Figure 2.12: Photo-peak efficiency simulated for different size of CeBr<sub>3</sub> detector.



Figure 2.13: Photo-peak efficiency simulated for different size of NaI(Tl) detector.



Figure 2.14: Comparison of photo-peak efficiency simulated for 3" x 3" LaBr<sub>3</sub>:Ce, CeBr<sub>3</sub> and NaI(Tl) detector.

## 2.7.4 Simulation of Response of LaBr<sub>3</sub>:Ce from Radioactive sources

The response of LaBr<sub>3</sub>:Ce, when exposed to radioactive sources, has been simulated using Geant4 simulation. Radioactive sources like <sup>137</sup>Cs, <sup>22</sup>Na, <sup>60</sup>Co used for this simulation. The radioactive source used are point source with isotropic in nature. The source is kept at a distance 20 mm from the detector surface. The response of these sources when expose to LaBr<sub>3</sub>:Ce detector is shown in Figure 2.15. From the simulation spectra it has been observed that when the detector is exposed to radioactive source, the gamma ray for the corresponding source have been generated using Geant4 simulation.







Figure 2.15: Response of a 3" x 3" LaBr<sub>3</sub>:Ce detector when exposed to radioactive sources. (a) <sup>22</sup>Na, (b) <sup>60</sup>Co and (c) <sup>137</sup>Cs.

Using GEANT4 simulations, the efficiency curve for LaBr<sub>3</sub>:Ce, CeBr<sub>3</sub> and NaI(Tl) detectors have determined. This simulation has been carried for different volumes of the detector. The results show that the efficiency of both LaBr<sub>3</sub>:Ce and CeBr<sub>3</sub> are much better than NaI(Tl). With  $3'' \ge 3''$  size of detector the efficiency at 8 MeV ~16% for LaBr<sub>3</sub>:Ce and CeBr<sub>3</sub> which is ~5% for NaI(Tl). We have simulated with the GEANT4 Monte Carlo the response of radioactive sources and could see all the energy lines from these source using LaBr<sub>3</sub>:Ce and CeBr<sub>3</sub>.

### Chapter - 3

## Development of a Cerium Doped Lanthanum Bromide Gamma Ray Spectrometer for Planetary Missions and Feasibility Studies

#### 3.1 Introduction

Most semiconductor and scintillator detectors can be used both at the laboratory as well as in space mission for gamma ray measurements. 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. Amongst semiconductor detectors, CZT (Cadmium Zinc Telluride) and High Purity Germanium (HPGe) have been used in the development of gamma ray spectrometers. High Purity Germanium (HPGe) detectors have been used in the 2001 Mars Odyssey Mission (Boynton et al 2004) Selene (Kaguya) (Hasebe, et al, 2008), and Chang'E-1 (Ma et al. 2008) for moon missions, and recently the MESSENGER (Goldsten, 2007) mission for probing
Mercury. 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. The FWHM (energy resolution) for this detector was 4.1 keV (~0.3%) at the 1332 keV line from <sup>60</sup>Co. High energy resolution HPGe detectors require cooling to liquid nitrogen temperatures (77 K). In space-based GRS these temperature is achieved by using a cry cooler for which ~20 W power is needed. The main disadvantage of these detectors is their poor energy resolution, and are not considered for the present experiment.

The discovery of Cerium doped lanthanum bromide (van Loef et al, 2002) in Delft University of Technology, provides a resolution of  $\sim 3\%$  at 662 keV (<sup>137</sup>Cs) and the advantage of this scintillation detector that it does not require cooling. In this chapter the physical properties of LaBr<sub>3</sub>:Ce will be discussed. The development of Gamma ray spectrometer and its performance with LaBr<sub>3</sub>:Ce detector will be discussed along with its suitability for the planetary application.

## 3.2 Cerium Doped Lanthanum Bromide Detector (LaBr<sub>3</sub>:Ce)

In search of an ideal scintillator, extensive research work has been carried out using oxide material and attractive oxide scintillators have been developed for medical and high energy physics applications in particular, LaBr<sub>3</sub>:Ce and LaCl<sub>3</sub>:Ce. The development of Ce doped LaBr<sub>3</sub> and LaCl<sub>3</sub> scintillators led to discovery of a new inorganic scintillators. LaCl<sub>3</sub>:Ce has several good qualities an inorganic scintillator should possess like high light yield (50, 000 photons/MeV), a fast decay (20 ns), good timing properties and an excellent energy resolution (3.2% @662 keV) (KS Shah et al. 2003). However, to achieve high light output and better resolution a relatively high Ce concentration was needed and the contribution of the short decay component to the total light yield was never to exceed 69% [van Loef 2000].

#### 3.2.1 Physical Properties of LaBr<sub>3</sub>:Ce Detector:

LaBr<sub>3</sub> crystal has a hexagonal (UCl<sub>3</sub> type) structure and a density of 5.29 g/cm<sup>3</sup> and melting point of 783° C. Because of low melting point of LaBr<sub>3</sub> it is possible to develop LaBr<sub>3</sub> crystals of pure LaBr<sub>3</sub> and cerium doped LaBr<sub>3</sub> by Bridgman technique. The detailed experimental technique is explained in Guillot-Noël, et al. (1999).

	LaBr <sub>3</sub> :Ce	NaI(Tl)	CsI(Tl)	BGO	CdZnTe	Ge
Density,	5.1	3.7	4.5	7.1	5.8	5.3
g cm <sup>.3</sup>						
Light out,	63	38	54	9	NA	NA
Photons per keV						
FWHM	3%	8%	8%	10%	5%	1%
at 662 keV						
1/e Decay	16	250	1000	300	NA	NA
Constant (ns)						
Thickness to stop	1.8	2.5	2	1.1	1.6	1.85
50 % of 662 keV						
Temp. coeff. Of light	0	-0.3	0.01	-1.2	NA	NA
output at 25º C						
Peak λ, nm	380	415	550	480	NA	NA

#### Table 3.1: Comparison of properties of gamma ray detectors.

The newly developed Ce-doped (5%) lanthanum halide scintillators LaBr<sub>3</sub>: Ce (E.V.D. van Loef, 2001) can be potentially used for gamma ray spectroscopy because of their better energy resolutions and proportionality of response as compared with traditional scintillating materials. LaBr<sub>3</sub>:Ce has a very fast light output decay (~16 ns) (< 10% of the NaI(Tl)) and an emission spectrum peaking in the blue end of the spectrum near 380 nm. The light yields are typically 65,000 photons/MeV or 160% relative to NaI(Tl). It provides a good energy resolution of ~3% at 662 keV (van Loef, 2001, 2000). Due to the high Z of lanthanum and high density of the crystal, a LaBr<sub>3</sub>:Ce detector with large volumes, can be potentially used for high-energy  $\gamma$ -rays (up to 20 MeV). Comparison of some of the characteristic features of LaBr<sub>3</sub>:Ce with commonly used scintillation detector as well as with some widely used solid state detector has shown in Table 3.1.

#### 3.2.2 Mechanism of Scintillation

The scintillation process in LaBr<sub>3</sub> involves two steps of mechanisms. A fast mechanism occurs almost immediately due to the trapping of free holes and electrons by Cerium, which is a temperature independent process. Next, a thermally activated process occurs where electrons and holes are trapped by a Self-Trapped Exciton (STE) fallowed by a subsequent energy transfer to Cerium. This process is dependent on the Cerium concentration and decreased as the crystal temperature is increased beyond 100 K.

#### **Hole formation**

#### **STE Formation**

 $Br^{-1} \rightarrow Br^{0} + e^{-} \qquad LaBr^{+} + Br_{2}^{-1} \rightarrow LaBr_{3}^{+}$  $Br^{-1} + Br^{0} \rightarrow Br_{2}^{-1} \qquad LaBr^{+} + Br_{2}^{-1} \rightarrow LaBr^{+} + Br_{2}^{+}$ 

#### **Electron Trapping**

 $La^{+3} + e^- \rightarrow La^{+2}$  $La^{+2} + Br^{-1} \rightarrow LaBr^+$  Many recent scintillations have been developed using Ce doped crystals. Out of these the light yield of Ce doped Lanthanum bromide (LaBr<sub>3</sub>:Ce) detector is more than other Ce doped scintillation crustal. The light output of the LaBr<sub>3</sub>:Ce also affected by the concentration of Ce doping. The light yield is maximum at 0.5% Ce doped crystal and is decreased with increase in doping concentration (van Leof et. al. 2002). The scintillator crystals light yield also affected by the change in temperature. Literature study shows that the light output of LaBr<sub>3</sub>:Ce is varied by 1% when the temperature varies from 0 to 55° C, while it changes by 5% when the temperature varies from -65° C to 140° C (Saint Gobain Data sheet). At room temperature the light output of LaB<sub>3</sub>:Ce is 160% s compared to that of NaI(TI) detector.

#### 3.2.3 Intrinsic Activity of LaBr<sub>3</sub>:Ce

It is well known that LaBr<sub>3</sub>:Ce, is an excellent scintillator with high light outputs (65,000 photons/MeV) and a very good energy resolution (~3% FWHM at 662 keV, <sup>137</sup>Cs). At the beginning, LaBr<sub>3</sub>:Ce crystals have only been available in small size due to the crystal anisotropic characteristics. In fact, LaBr<sub>3</sub>:Ce shows strong anisotropy in thermal expansion, heat transfer and mechanical strength hence its larger ingots are likely to crack during the cool down process after the growth. Now a day with improvement in technique large size crystals are grown.



Figure 3.1: Decay scheme of <sup>138</sup>La.



Figure 3.2: Self-counting background spectrum of LaBr<sub>3</sub>:Ce detector.

In natural lanthanum, the isotopic abundances of <sup>139</sup>La and <sup>138</sup>La are about 99.91 % and 0.09% respectively shown in Figure 3.1. Radioactive <sup>138</sup>La decays to stable isotopes by beta decay (33.6%) and electron capture (66.4%). <sup>138</sup>La decays to <sup>138</sup>Ce by beta decay with 789 keV gamma energy; further, <sup>138</sup>La decays to <sup>138</sup>Ba by capturing an electron emitting gamma energy of 1436 keV. The refilling of electron shell results in an emission of barium x-ray energy of 32 KeV. The gamma spectrum of self-counting of LaBr3:Ce is shown in Figure 3.2

# 3.3 Feasibility Studies of LaBr<sub>3</sub>:Ce using Commercial Setup

Our feasibility studies used a 3" diameter x 3" length LaBr<sub>3</sub>:Ce from Saint Gobain, where LaBr<sub>3</sub>:Ce is coupled to a 3" inch PMT (photo-multiplier). The PMT output is coupled to pre-amplifier and voltage divider, from Saint Gobain. The output of pre-amplifier further amplified by Ortec 671 NIM Module. The detector PMT has been biased through a Canberra HV 3106D NIM module. The measurement was carried out at a peaking time of 2 µs. The Gaussian shape

amplified output from the amplifier is then digitized using the Ortec 8K multichannel analyzer (MCA). The digital output from the MCA is readout using Ortec Maestro software. For all measurements, the detector was kept inside a 4-inch lead shield chamber.

#### 3.3.1 Calibration and Energy Resolution

The conversion of measured ADC channel to energy requires analysis of gamma ray spectra collected and fitting with Gaussian distribution functions for estimation of peak centroid and FWHM. The energy calibration and linearity measurement of the LaBr<sub>3</sub>:Ce detector has been carried out using a number of radioactive sources (<sup>22</sup>Na, <sup>133</sup>Ba, <sup>137</sup>Cs, <sup>57</sup>Co, <sup>60</sup>Co, <sup>108</sup>Cd, <sup>241</sup>Am, <sup>152</sup>Eu, and <sup>226</sup>Ra). Pulse height spectra have been recorded (Figure 3.3) for each calibration source which are kept at a distance of 10 cm and are counted for 300 seconds.











Figure 3.3: Response of LaBr<sub>3</sub>:Ce to different radio-active sources, (a) <sup>214</sup>Am, (b) <sup>133</sup>Ba, (c) <sup>137</sup>Cs, (d) <sup>22</sup>Na, and (e) <sup>152</sup>Eu.



Figure 3.4: Linear curve of ADC Channel vs Energy for LaBr<sub>3</sub>:Ce.



Figure 3.5: Energy resolution achieved at different energies for LaBr<sub>3</sub>:Ce detector.

The peak height of each energy region was calculated by fitting the photopeak areas of these energies to a Gaussian function. The sigma ( $\sigma$ ) was also obtained using fitting and the energy resolutions of the each energy were calculated. The ADC channel corresponding to the peak height of each energy is plotted and is shown in Figure 3.4. The ADC channel and the energy are correlated linearly with a slop of 0.39 and intercept 4.65.

The energy resolutions for each energy were estimated with the following formula,

$$R = \frac{2.35 \ x \ \sigma}{Peak \ Height} \ x \ 100 \ \%$$

Where, sigma ( $\sigma$ ) and the peak height of the corresponding energy are obtained from the Gaussian fittings. The gamma response shows a decay curve and the variation of energy resolution as shown in Figure 3.5 can be fitted using the following equation.

$$R = 103.23 \ x \ E^{-0.55}$$

#### 3.4 Development of Gamma Ray Spectrometer

Presently, the laboratory model of the gamma ray spectrometer (GRS) has been developed and subsequent developments would focus on space-borne experiments. We have attempted to develop the full readout system using FPGA based processing electronics.



Figure 3.6: Block diagram of LaBr3:Ce gamma ray spectrometer.

In the past, some instruments are developed using controller and ASCI, the realization of DC signals are much better-using FPGA and ADCs. FPGA provides an opportunity to realize analog as well as digital circuits in a more convenient way through the logical algorithm. The laboratory model is developed using commercial grade components, which are also available in space-qualified grade. The block schematic of the LaBr<sub>3</sub>:Ce based gamma ray spectrometer is shown in Figure 3.6.

The development design of GRS is mainly divided into five parts

- (i) Detector
- (ii) Biasing Module,
- (iii) Front-End-electronics
- (iv) Data processing

The details of each subsystem are described in the following sections.

### 3.4.1 Detector Configuration



Figure 3.7a: 3" x 3 " LaBr<sub>3</sub>:Ce detector Figure 3.71b: Front view of couple with a 3.5" XP-5700 PMT.

voltage divider & preamplifier.

Table 3. 2 : Prpoerties of LaBr3:Ce.

Crystal	LaBr <sub>3</sub> :Ce
Size	3 inch diameter x 3 inch
	height
Light Yield	65,000 ph/MeV
Decay Time	16 ns
Light output	380 nm
Energy resolution	2.8 % @ 662 keV
Energy range	0.1 to 10 MeV
Operating temperature	0 to 250 C

Model	XP-5700
Size	89 mm round tube
Dynode	with 10-stage
Glass Window	Borosilicate glass
Cathode	Bi-alkali photo cathode
Refractive index	1.54 at 420 nm (spectra range 270-
	650nm).
Operating	0 to 250 C
temperature	

Table 3.3: Properties of PMT coupled to LaBr<sub>3</sub>:Ce detector

The present LaBr<sub>3</sub>:Ce (BrilLanCe 380) as shown in Figure. 3.7a and 3.7b detector from Saint Gobain is a 3" diameter with 3" height coupled with a 3.5-inch Photonics 10 stage XP5700 PMT. Physical properties of LaBr<sub>3</sub>: Ce and the coupled PMT are given in Table 3.2 and 3.3. For the charge to be carried out to the next dynode and finally convert the charge to current, a suitable bias voltage was applied to the PMT. The PMT required a biasing voltage of approximately 720 V for its optimum performance. We have used a Canberra high voltage NIM module (module No 3106D) for generating 720 V DC bias voltage.

#### **3.4.2 Front End Electronics**

The front-end electronics of the gamma ray spectrometer consists of charge sensitive pre-amplifier (CSPA), shaping amplifier, event triggering, peak detector, analog to digital converter (ADC). The details of these subsystems are described below,

#### 3.4.2.1 Charge Sensitive Pre-Amplifier (CSPA)

The CSPA can be either of RC or reset type. Presently RC type, from Saint-Gobain, model number AS-2612, has been used in this work. The output of the preamplifier is a negative ramp output with an amplitude of few hundred millivolts to  $\sim 500$  mV and a pulse width of 100 ns rise time with a decay time of 50  $\mu$ s. The output from pre-amplifier captured with Tektronix Oscilloscope is shown in Figure 3.8.



Figure 3.8: Snapshot of pre-amplifier output of LaBr<sub>3</sub>:Ce, captured with Tektronix Oscilloscope detector.

#### 3.4.2.2 Shaping Amplifier

For pulse-height or energy spectroscopy, the linear pulse shaping amplifier carries out several vital functions. Its prime role is to increase the amplitude of the preamplifier output pulse from the mV range into the  $0.1 \sim 10$  V range. For the accurate pulse height measurements necessitate a peak-sensing analog-to-digital converter (ADC). Furthermore, the amplifier shapes the pulses to optimize the energy resolution, and to minimize the possibility of overlap between consecutive pulses. For most radiation detectors, achieving the optimum energy resolution requires long pulse widths. On the other hand, short pulse widths are necessary for high counting rates. In such cases, a compromise pulse width must be selected

so that the spectroscopy system can be optimized. Various techniques available for pulse shaping in the linear amplifier are,

- Single Delay Line (SDL)
- Double Delay Line DDL
- CR-RC method

In the present design, we have opted is for CR-RC technique which is described below.

**CR-RC** method



Figure 3.9: Schematic of CR-RC Network.

The preamp signal first passes through a CR shaper and then RC shaper. The CR network works as a differentiator or a high pass filter and the RC network works as integrator or a low pass filter as shown in Figure 3.9. *For CR network* 

$$E_{out} = \tau \frac{dE_{in}}{dt}$$

Where,  $\tau$  = RC is time constant.

For RC network

$$E_{out} = \frac{1}{\tau} \int E_{in} dt$$

Where,  $\tau$  = RC, time constant.

The time domain and frequency domain response of CR, RC and CR-CR are shown in Figure 3.10



Figure 3.10: The time and frequency domain response of CR, RC and CR-RC.



Figure 3.11: Block schematic of shaping amplifier.

The block diagram schematic of the breadboard model for the shaping amplifier has been designed with a three-stage amplification i.e. CR-RC-RC (CR-RC<sup>2</sup>) with five-pole amplification as shown in Figure 3.11. The shaping amplifier has been designed using AD829 a very low noise, high speed, wide bandwidth video amplifier (AD-829 Data Sheet). Four-layer PCB has been fabricated for the breadboard model of the shaping amplifier using AD829. The 1<sup>st</sup> stage of the shaping amplifier consists of CR-high pass filter which basically acts as a differentiator. The 1<sup>st</sup> amplification stage is adjusted to a gain of 2. Both the 2<sup>nd</sup> and 3<sup>rd</sup> stage shaping amplifier are RC-low pass filters, and the gain values for these stages are set to 2 and 1.2 respectively. The 3<sup>rd</sup> stage of shaping amplifier is

kept in an adjustable mode to test the final shaping at different voltage level upto a maximum level of 10 Volts. The shape of the output at the 3<sup>rd</sup> amplification stage is a Gaussian shape. The total gain of the breadboard model of the shaping amplifier has been set to a gain of ~4.8. With the present gain setting, the output pulse corresponds to a maximum energy of ~3 MeV (At present set to measure upto  $^{208}$ Tl i.e. 2.615 MeV). For the space instruments, the energy range can be set to 10 MeV where it can measure high energy Fe and Al elements.

The shaping amplifier module has been designed for both positive and negative pre-amplifier output. The selection of positive and negative signal can be done through jumper settings. For the negative preamplifier output, the signal will be first inverted with a unit gain and the output of the inverter is connected to the 1<sup>st</sup> stage of the shaping circuit (CR-RC<sup>2</sup>), while the positive pre-amp signal will be directly connected to the shaping circuit.

In CR-RC circuit the output pulse undershoots when it attempts to return to the baseline. This happens because of the long exponential decay on the preamplifier output pulse. Thus the apparent pulse amplitudes measured for these pulses will be too low, which leads to a broadening of the peaks recorded in the energy spectrum. To eliminate the undershoot a pole-zero cancellation circuit has been incorporated. To adjust the pole-zero of the signal, an additional resistance has been used across the capacitor of the 1<sup>st</sup> amplifier (CR-amplification stage) as shown in Figure 3.11. The pole-zero cancellation also improves the peak shapes and resolution in the energy spectrum at high counting rates.

To achieve a good energy resolution and peak position stability at high count rate, the amplifiers are entirely dc-coupled connected (except for the CR differentiator network). As a consequence, the dc offsets of the earliest stages of the amplifier are magnified by the amplifier gain and it causes a large and unstable dc offset at the amplifier output. To nullify this dc offset a baseline restorer (BLR1 from Amptek) has been used soon after the pole-zero as shown in Figure 3.11. The output from pre-amplifier, inverter and different stage of the shaping amplifier have been captured in oscilloscope and are shown in Figure 3.12 and 3.13.



Figure 3.12: Snapshots of different stage of shaping amplifier output captured with Tektronix Oscilloscope.



Figure 3.13: Output at different stage of shaping amplifier. amplifier output.

#### **3.4.3 Processing Electronics**

#### 3.4.3.1 Event Triggering

To record the event accurately the shaping amplifier output is passed through the event trigger circuit to generate digital pulses for each  $\gamma$ -pulse interacting with the detector to initiate the digital system for data processing. The event trigger has been designed with a fast voltage comparator which provides pulse output when the input signal amplitude crosses the set threshold. The set threshold voltage also acts as a LLD for the spectrometer.

#### 3.4.2.2 Peak Detector

A peak detector detects the peaks of an input pulse or waveform and produces an output based on the detected peaks. The output of the peak detector mainly depends on the type of peak detector used in FEE. Out of the various type of peak detectors some peak detector produces a digital output containing information about when positive and negative peaks of a pulse or waveform occur. While some other peak detectors produce an analog output with a magnitude equal to the last detected peak, or the magnitude of the maximum peak encountered. Accurate detection of the peaks for an input pulse or waveform can be useful in a variety of different applications. The basic block schematic of the peak detector is shown Figure 3.14.



Figure 3.14: Block schematic of peak detection.

When the capacitor is shorted (Reset) and if there is a positive voltage at the non-inverting input, the output voltage of the op-amp forward biases the diode and charges up the capacitor. The charging of the capacitor last until the inverting and non-inverting inputs are at the same voltage, which is equal to the input voltage. When the non-inverting input voltage exceeds the voltage at the inverting input, which is also the voltage across the capacitor, the capacitor will charge up to the new peak value. Consequently, the capacitor voltage will always be equal to the maximum positive voltage applied to the non-inverting input. Once charged, the peak detected is stored upto certain time, which mainly depends on the impedance of the load connected to the circuit. Subsequently, the capacitor will slowly discharge towards zero. A voltage follower can be used to minimize the discharge rate. The voltage follower buffer the detector's output from an external load, momentarily shorting the capacitor to ground will immediately set the output to zero.



Figure 3.15: Bock schematic of PH-300 peak detector, (adopted from Amptek data sheet).



## Figure 3.16: Different mode of operation of PH-300 (adopted from Amptek data sheet).

The detection of the peak of the Gaussian-shaped pulse forms the shaping amplifier has been carried out by using a PH300 peak detector manufactured by AMPTEK. PH300 peak detector is available in the hybrid form with high speed, lowest drop rate and consume very low power. The input signal to PH300 gated through a linear gate. The linear gate is controlled by a TTL compatible or digital logic signal. When the gate is open the error amplifier allow the input pulse. When the gate is closed, the error amplifier rejects the input pulses by grounding the input of the error amplifier. During the rise time of the input pulse, the hold capacitor is charged through a charging diode and a holding resistor, which is referred to as the charging mode. In the charging mode, the output voltage of PH300 follows the input signal and the peak-detector logic output is in the inactive state. Once the input pulse reached to its maximum voltage the device goes into hold mode and then it starts to decay. This state can be completed by discharging the hold capacitor. When the logic signal is activated to accomplish this, the PH300 is placed in discharge mode. A special case of the discharge mode is the tracking mode. The functional block schematic of PH300 is shown in Figure 3.15. The timing diagram for the different mode of operation is shown in Figure 3.16. The control logic required to operate the PH-300 is designed such that the peak detector holds the analog pulses for the duration of 5  $\mu$ s which include the completion of ADC conversion and 14-bit parallel data readout. We have used a 14-bit parallel ADC. The detailed functioning of these control signal will be discussed in the timing diagram.

#### **3.4.3.3** Analog to Digital Converter (ADC).

The peak detector output is digitized using an AD-7899 parallel ADC. AD-7899 is a fast low power consuming (16 mW power consumption) 14-bit (13 data bit and one bit for sign) ADC. AD7899 is having a 2.2  $\mu$ s standard successive approximation ADC with high throughput, a track/hold amplifier, 2.5 V reference, on-chip clock oscillator, signal conditioning circuitry, a high-speed parallel interface (400 KSPS). It can accept analog input ranges of ±10 V, ±5 V, ±2.5 V, 0 V to 2.5 V, and 0 V to 5 V. Therefore, this ADC allow us to have a wide range of input pulse and we can also use bipolar input signal.

The track/hold amplifier on the AD7899 allows the ADCs to accurately convert an input sine wave of full-scale amplitude to 14-bit accuracy. The track/hold amplifiers acquire input signals to 14-bit accuracy in less than 300 ns. The track/hold amplifier activate the sampling on the rising edge of CONVST (start of conversion) and when the conversion is completed it returns to tracking mode. The required 2.5V reference voltage can be provided either by internal supply

from  $V_{REF}$  or by an external 2.5 V reference. To optimize the ADC circuit internal reference has been used by connecting  $V_{REF}$  PIN to ground (AGND) through 0.1  $\mu$ F capacitor. The conversion is initiated by applying a rising edge to the CONVST signal. The conversion time depends on the clock used. The AD7899 has an internal laser trimmed and also an external clock source can be used to control the conversion process. But to optimize the space and to have a fast conversion internal clock has been used. The status of the conversion is indicated by the dual function signal BUSY/EOC. The V<sub>DRIVE</sub> feature controls the voltage levels on the I/O digital pins. The digital data are being processed by an FPGA.

#### 3.4.3.4 FPGA Based Data Acquisition System



Figure 3.17: Block diagram of interfacing of different control signals from FPGA to Peak detector, ADC and interfacing DAQ (data acquisition)

To control proper functioning of the peak-detector and ADC conversion an A3P-250 FPGA manufactured by Actel has been used. The control logic signal required for the peak detector and the ADC are generated by FPGA. The programming of the FPGA has been done through VHDL code. The interfacing of these signals with the FPGA, Peak detector, and ADC is shown in the block diagram as in Figure 3.17. The digitized data from ADC are fed to FPGA through line driver. The breadboard module of processing board is shown in Figure 3.18.



Figure 3.18: Breadboard module of processing electronics for gamma ray spectrometer.

The digitized data from ADC are processed by a FPGA based data processing module. Through a line driver, these digital data are interfaced to a NI (National Instrumentation) PCIe 6537 digital I/O data acquisition module. The PCIe data acquisition module is interfaced to the computer through LabVIEW. Using LabVIEW, a data acquisition software has been developed for data fetching and converting these data into a spectrum form and for further analysis. A screen snapshot of the data acquisition software is shown in Figure 3.19.

#### 3.4.4 Data Readout System using LabVIEW

The gamma ray spectrometer is designed such that the system detects every event after 5  $\mu$ s from the previous event. Within 5  $\mu$ s the pulse generated by the detector are rejected. The detailed processing of the pulse signal from shaping amplifier upto the data fetching using data acquisition software can be explained with the timing diagram as shown in Figure 3.23. First, the pre-amplifier pulse is amplified with the CR-RC<sup>2</sup> shaping amplifier in the form of Gaussian shaped output. Along with the shaping amplifier, the event triggered pulse is also generated, which indicate a gamma pulse has been detected. On the rising edge of the event triggered pulse, the peak detector starts charging to its maximum by activating the GATE and RAMP signals. The GATE and RAMP signals are generated by FPGA. Once the peak detector reaches its maximum, PKDT is activated by a negative edge signal and then the GATE is deactivated so that the subsequent signals can be blocked for the next 5  $\mu$ s. With falling edge of the Gate, the ADC is initiated to start conversion by activating the CS\_Bar.



### Figure 3.19: Snap shot of data acquisition software developed using LabVIEW.

On the activation of CS\_BAR, the start of conversion (CONVST\_BAR) is also activated to start the conversion. Once the conversion is completed the ADC provides a pulse through EOC (End of Conversion). On the falling edge of the EOC pulse, the 14-bit data are fetched by FPGA through the line driver. Then the digitalized data from the FPGA are provided to DAQ card. Along with the 14-bit digital data, a reference (REF) pulse is also provided to the data acquisition module which synchronizes with the falling edge of RAMP pulse of the peak detector. This reference pulse is connected to the clock output strobe of the data acquisition module. When the start button of the acquisition software is activated the data acquisition module checked for the rising edge of clock synchronization pulse connected to its clock output strobe. One the rising edge of the clock synchronization pulse data fetching from FPGA to the computer is started.



Figure 3.20: Timing diagram for data processing of gamma ray spectrometer.

#### 3.5 Results

#### 3.5.1 Experimental Setup

To verify the performance gamma ray spectrometer developed, a 3"  $(diameter) \times 3$ " (thickness) LaBr<sub>3</sub>:Ce crystal (BrilLanCe 380) coupled to a 3.5-inch photomultiplier (PMT), voltage divider and pre-amplifier from Saint Gobain has been used. To improve the signal-to-noise ratio, a single breadboard has been designed where the design of shaping amplifier, discriminator, peak-detector, ADC, and FPGA are incorporated. The breadboard model is shown in Figure 3.21. The required control logic signals were generated through a FPGA kit and digital

data from FPGA processing board were readout through NI (National Instrumentation) data acquisition card, which is interfaced to the computer by data acquisition software developed in LabVIEW. To reduce the background, all measurements were carried out within a lead shield chamber of a wall thickness of 4".



Figure 3.21: Laboratory Experimental setup.

#### 3.4.2 Calibration and Energy Resolution

The performance of the breadboard model was checked using a <sup>22</sup>Na radioactive source. The calibration and photo-peak identification have been done using a <sup>22</sup>Na radioactive source, and gamma ray lines arising from the intrinsic activity of <sup>138</sup>La as shown in Figure 3.22. The proportionality response of ADC channel with gamma ray energy for the LaBr<sub>3</sub>:Ce gamma ray spectrometer (i.e., the linearity curve) is shown in Figure 3.23. It can be observed that the ADC

channel and their corresponding energy line shows a good correlation and is fitted using a straight line with a slope (gain) of ~0.36 and an intercept (offset) of ~16. We have also evaluated the FWHM energy resolution at various energies for our LaBr<sub>3</sub>:Ce gamma ray spectrometer. The energy resolution at 511 and 1274 keV, measured using a <sup>22</sup>Na radioactive source, are 4.1% and 2.5% respectively (Panda et. al. 2016). Arnold et al (2012) have reported energy resolutions of 5.3% and 8.2% at 1461 and 2615 keV from "raw" gamma ray spectra uncorrected for <sup>138</sup>La intrinsic activity. Quarati et al. (2007) measured the energy resolutions of 2.8% and 3% at 662 keV for a 1″ × 1″ and 2″ × 2″ LaBr<sub>3</sub>:Ce gamma ray spectrometers.



Figure 3.22: Calibration of LaBr<sub>3</sub>:Ce using <sup>22</sup>Na radioactive source.



Figure 3.23: Calibration curve obtained using <sup>22</sup>Na radioactive source and internal activity of LaBr<sub>3</sub>:Ce.

Once the breadboard performance has been tested successfully with a single radioactive source, a number of radioactive sources used to calibrate the system for a wide range. Therefore, different radioactive sources (<sup>241</sup>Am, <sup>57</sup>Co, <sup>133</sup>Ba, <sup>109</sup>Cd, <sup>22</sup>Na, <sup>137</sup>Cs, <sup>60</sup>Co, <sup>152</sup>Eu, and <sup>226</sup>Ra) have been used in the next set of experiments. The response of these sources are shown in Figure 3.25. The raw data of each spectrum were smoothed using the moving average method. In order to extract the photo-peak centroid, the ROI of the peak has been identified from the measured spectra and then fitted using a Gaussian function. The peak centroid (Peak height) and the sigma (standard error) were obtained and used for the calibration of ADC channel to gamma ray energy, and to estimate FWHM. The peak height is then plotted against the corresponding energy line as shown in Figure 3.24. It is observed that the peak heights are fitted linearly with the energy lines with a slope of 0.39 and intercept of 10.38.



Figure 3.24: Calibration curve obtained using different radioactive source.









Figure 3.25: Gamma ray spectra measured with LaBr<sub>3</sub>:Ce using different radio-active source. (a) <sup>241</sup>Am, (b) <sup>133</sup>ba, (c) <sup>137</sup>Cs, (d) <sup>22</sup>Na and <sup>152</sup>Eu.



Figure 3.26: Energy resolutions with varying energy for LaBr<sub>3</sub>:Ce detector.

The energy resolution values are calculated from the sigma ( $\sigma$ ) obtained from the Gaussian fitting. The energy resolution obtained from the experimental results are plotted against their energy and compared with the theoretical energy resolution (1/ $\sqrt{E}$ ). Figure 3.26. shows an exponential decay in energy resolution with gamma energy and can be expressed as:

 $R(\%) = 112.3 \ x \ E^{-0.55}$ 

Few energy resolutions obtained from the breadboard model has been compared with that obtained from the commercial set up. The comparison is shown in Table 3.4. Table 3.4: Comparison of energy resolution achieved by GRS developed with Commercial electronics and in-house.

Parent	Energy	From	In-house	
Nuclide	keV	Commercial	developed	
		Electronics	GRS	
<sup>133</sup> Ba	81	11.0	11.2	
<sup>152</sup> Eu	121.8	7.6	7.8	
<sup>152</sup> Eu	244.7	5.0	5.3	
<sup>133</sup> Ba	356	3.8	3.9	
<sup>22</sup> Na	511	3.4	3.6	
<sup>226</sup> Ra	609	3.1	3.5	
<sup>137</sup> Cs	661.7	2.9	2.6	
<sup>22</sup> Na	1274	2.3	2.4	
<sup>60</sup> Co	1332	2.3	1.8	

#### 3.5.3 Background Measurement and Intrinsic Activity

The performance of LaBr<sub>3</sub>:Ce gamma ray spectrometer (GRS) has been tested and compared with NaI(Tl) detector. The background measurement was integrated for a long time period (~72 hrs). The background spectra measured with the breadboard model is shown in Figure 3.27a. From the Figure, it can be observed that the internal activity of LaBr<sub>3</sub>:Ce can be categorized on the basis of energy windows i.e. in the energy range 20 keV to 1600 keV and 1700 to 2800 keV. The internal activity in the energy range of 20 to 1600 keV is due to the presence of radioactive isotope of <sup>138</sup>La and in the internal activity in the energy range of 207Ac as contamination.



Figure 3.27a: Gamma ray background spectra obtained using LaBr<sub>3</sub>:Ce gamma ray spectrometer.

Natural Lanthanum contains 0.09% of radioactive <sup>138</sup>La ( $t_{1/2} = 1.05 \times 10^{11}$  y) which decays to produce an excited <sup>138</sup>Ce\* during 33.6% of its decays by beta emission, and the remaining 66.4% decays result in a production of excited <sup>138</sup>Ba\* by electron capture. <sup>138</sup>Ba\* decays by emission of a gamma energy of 1436 keV with a coincident 32 keV X-ray from Ba. Excited <sup>138</sup>Ce nuclei decay follows by the emission of 789 keV gamma ray in coincidence with the beta continuum having an end-point energy of 255 keV. From Figure 2.34b, a pronounced photo-peak is observed in the gamma spectrum at ~1436 keV with a coincident peak at 32 keV. A Compton continuum from the 789 keV which extends upto ~1040 keV due to  $\beta$ continuum was also observed. Other than these two peaks, few small peaks were observed from the background spectra within 1700 to 2800 keV energy range as shown in Figure 3.27b. These peaks are produced from the daughter products by alpha-decay from <sup>227</sup>Ac which basically comes as contamination during the process of crystal growth. Alpha decay of <sup>227</sup>Ac produces <sup>227</sup>Th, <sup>223</sup>Ra, <sup>219</sup>Rn, <sup>215</sup>Po, and <sup>211</sup>Bi. The peak position of these daughter elements are marked in Figure 3.27c.



Figure 3.27b: Background spectra of LaBr<sub>3</sub>:Ce measured upto 3 MeV. The plot shows peaks at 789 keV and 1436 keV intrinsic activity of <sup>138</sup>La.



Figure 3.27c: Presence of intrinsic activity in the energy range of 1.7 – 3 MeV within LaBr<sub>3</sub>:Ce detector due to <sup>227</sup>Ac.
The intrinsic activity count-rate for the LaBr3:Ce using the breadboard model of gamma ray spectrometer was calculated by integrating the counts from 20 to 3000 keV. Then the intrinsic activity of the <sup>40</sup>K, <sup>238</sup>U and <sup>232</sup>Th were calculated by integrating the counts in the energy region of 1400 -1520 keV, 1700 – 1800 keV and 2550 – 2700 keV respectively. The intrinsic activity was found to be 61 counts s<sup>-1</sup> (i.e.  $\sim$ 0.18 counts s<sup>-1</sup> cm<sup>-3</sup>) for the <sup>40</sup>K energy window (1400-1520) keV). This value is significantly higher than the value of 177 count minute<sup>-1</sup> ( $\sim 0.07$ counts s<sup>-1</sup> cm<sup>-3</sup>) for a smaller  $1.5'' \times 1.5''$  LaBr<sub>3</sub>:Ce detector reported by Arnold et.al., 2012. For the <sup>238</sup>U (1700-1800 keV) and <sup>232</sup>Th (2550-2700 keV) energy windows, the intrinsic activity counts rates for our detector are  $\sim 0.37$  counts s<sup>-1</sup> (0.0011 counts s<sup>-1</sup> cm<sup>-3</sup>) and  $\sim$ 3.4 counts s<sup>-1</sup> (0.01 counts s<sup>-1</sup> cm<sup>-3</sup>) respectively (Panda et. al. 2016). A comparative Table has been prepared to show the comparison of intrinsic activity calculated from both commercial set up and the breadboard model. The comparison of background count rates obtained from gamma ray spectrometer developed using commercial electronics and the GRS developed in-house is shown in Table 3.5. It is observed that the background count rates obtained from both electronic systems are almost same between 1.6 to 3.0 MeV.

Energy Commercial Set-up		In-house developed		
		bi eauboai u mouei		
		Counts s <sup>-1</sup> cm <sup>-3</sup>		
20-3000 keV	0.77	0.87		
<sup>40</sup> K	0.18	0.18		
<sup>238</sup> U	0.001	0.001		
<sup>232</sup> Th	0.009	0.01		

Table 3.5: Comparison of intrinsic activity measured from GRS developedwith commercial electronics module & the module developed in house.

## 3.5.4 Estimation of K, U & Th Concentration in Terrestrial Samples

As a first attempt towards realizing our scientific objectives, we have investigated the feasibility of using a LaBr<sub>3</sub>:Ce gamma ray spectrometer for determination of K, U and Th concentrations in soil/rock samples. The concentration of these elements can be measured using gamma ray produced by the decay of <sup>40</sup>K for K, <sup>238</sup>U for U and <sup>232</sup>Th for Th. <sup>40</sup>K decays to <sup>40</sup>Ar with an emission of gamma energy 1461 keV. The prominent gamma energy lines that can be used for measurement of U are 1764 keV(<sup>214</sup>Bi), 1120 keV(<sup>214</sup>Bi), 609 keV (<sup>214</sup>Bi) and 352 keV(<sup>214</sup>Pb). Similarly, <sup>232</sup>Th decays and emits gamma lines at 2615 keV(<sup>208</sup>Tl), 911 keV (<sup>228</sup>Ac), 583 keV (<sup>208</sup>Tl), and 239 keV (<sup>212</sup>Pb). For <sup>40</sup>K, 1400 - 1520 keV energy window, for <sup>238</sup>U 1700 – 1800 energy window and for <sup>232</sup>Th 2550 – 2700 keV have been chosen (Arnold et. al. 2012).





Figure 3.28: Comparison of gamma ray spectrums measured with LaBr<sub>3</sub>:Ce from background and samples (a) US#110 and (b) BBS#2.

We have investigated the feasibility of using a LaBr<sub>3</sub>:Ce gamma ray spectrometer for determination of Th, K and U concentrations in soil samples. US-110 is a rock powder standard of basaltic composition prepared in PRL about 30 years ago. BBS-2 is a red soil collected from southern India. The K, U and Th values for US#110 and BBS#2 have been determined independently using a HPGe gamma ray spectrometer and the results are shown in Table 3.6. The gamma ray spectrum of USD#110 is integrated over 24 hrs. and is shown in Figure 3.28. The measurement from HPGe for BBS#2 shows that the concentration of U and Th quite higher than the normal soil/rock, therefore the gamma ray spectrum for BBS#2 has been acquired by integrating for 2hr and is shown in Figure 3.28. The background spectra are shown as a comparison for in both US#110 and BBS#2. Using the 1400-1520 keV, 1700-1800 keV and the 2550-2700 keV energy windows, we attempted to estimate the K, U and Th concentrations of the sample US#110, using KCl as a standard for K and BBS#2 as standards for U and Th. However, due to large intrinsic activity of our LaBr<sub>3</sub>:Ce crystal, we were unable to estimate the concentrations of Th, U and K. Next, we attempted to determine the K, U, and Th concentrations of US#110 using a NaI (Tl) gamma ray spectrometer,

connected with front-end and processing electronics developed as described above.

Sample	Weight	К	U	Th	
	gm	%	ppm	ppm	
US#110	51.85	0.78	2.91	13.2	
BBS#2	45.47	1.56	48	1515.3	

 Table 3.6: Concentration of US#110 & BBS#2 measured with HPGe.

#### 3.5.5 Comparative Study of LaBr<sub>3</sub>:Ce and NaI(Tl)

This section describes the comparison of LaBr<sub>3</sub>:Ce detector and NaI(Tl) detectors for gamma ray spectroscopy and U, Th and K estimation in samples. NaI(Tl) is a commonly used scintillation detector used for radiation measurements. The energy resolution of NaI(Tl) is around 6-7 % at 662 keV, while it is <3% at 662 keV for LaBr<sub>3</sub>:Ce. The light output of NaI(Tl) is 1.5 less than LaBr<sub>3</sub>:Ce (48000 per MeV), and the decay time of NaI(Tl) is ~300 ns. But as the intrinsic activity is very low as compared to LaBr<sub>3</sub>:Ce, it is possible to estimate the concentration of K, U, and Th as compared to LaBr<sub>3</sub>:Ce.

A 2" × 2" NaI (Tl) detector from Saint Gobain has been used in our studies. To reduce background levels, all measurements for NaI detector were carried out inside a 4-inch-thick lead shield chamber. The detector is calibrated using <sup>22</sup>Na radioactive sources. The response and calibration curves obtained using <sup>22</sup>Na are shown in Figure 3.29 and 3.30 respectively. For the electronics system developed in-house, the energy resolution was observed to be 4.1% @1274 (<sup>22</sup>Na) for NaI(Tl), (Panda et. al. 2016).



Figure 3.29: Response of NaI(Tl) from <sup>22</sup>Na radioactive source.



Figure 3.30: Calibration curve of NaI(Tl) detector.

The measurements using  $2'' \times 2''$  NaI (Tl) detector showed much lower background measurement values for the above energy windows mentioned in the

earlier section, and consequently, a correction for internal radioactivity is not essential in the analysis of K, Th and U concentrations. The background spectra measured using NaI(Tl) detector is shown in Figure 3.31. The internal radioactivity contributions as obtained from self-counting during a background measurement are ~ 0.0014, 0.00008 and 0.00006 counts s<sup>-1</sup> for the K, U and Th energy windows respectively. A comparison of the internal activity of NaI(Tl) and LaBr<sub>3</sub>:Ce has been given in Table 37. Form Table 3.7it can be realized the intrinsic activity in the range of 20 -3000 keV is nearly two orders lower than that of LaBr<sub>3</sub>:Ce. In the K, U and Th windows the intrinsic activity are also two orders lower than that of LaBr<sub>3</sub>:Ce.



Figure 3.31: Background spectra measured with 2" x 2" NaI(Tl) detector.

Table 3.7: Comparison of Intrinsic activity from LaBr<sub>3</sub>:Ce & NaI(Tl).

Detector	20-3000 keV	<sup>40</sup> K	<sup>238</sup> U	<sup>232</sup> Th
	Counts s <sup>-1</sup> cm <sup>-3</sup>			
LaBr <sub>3</sub> :Ce	0.87	0.18	0.001	0.01
(3″ x 3″)				
Na(Tl)	0.01	0.0014	0.00008	0.00006
(2″ x 2″)				



Figure 3.32a: Gamma ray spectra from US#110 measured with NaI(Tl).



Figure 3.32b: Gamma ray spectra from BBS#2 measured with NaI(Tl).

Table 3.8: Elemental Concentrations of samples determined using HPG	e
and NaI(Tl) gamma ray spectroscopy.	

Sample	HPGe				NaI(TI	)
	K	U	Th	K	U	Th
US#110	0.78	2.01	12.2	0.87		11 /
05#110	0.78	2.91	13.2	0.07	-	11.4
GHRF#2	3.38	14.7	48	3.31	17.92	41.65

Next, we attempt to estimate K, U and Th concentration for any geological sample (rock/soil) using the in-house developed gamma ray spectrometer. For this purpose, BBS#2 and US#110 have been used, as described above. In this study, BBS#2 was used as the standard for Th estimation (BSS#2, Th = 1515 ppm). The spectra from US#110 and BBS#2 are shown in Figure 3.32a and 3.32b. As the internal radioactivity of a NaI (Tl) crystal is negligible in comparison to LaBr<sub>3</sub>:Ce (Figure 3.31), the concentration of Th and K could be estimated using the 1461 and 2615 keV gamma ray lines. The Th concentration of US#110 was estimated to

be ~11.4 ppm. and is within 14% of the 13.2 ppm value determined using a HPGe gamma ray spectrometer. The K concentration of US#110 was estimated to be 0.87%, and is within ~10% of the 0.78% value determined using a HPGe gamma ray spectrometer (Panda et al. 2016). U estimation was not feasible in the present study using NaI (Tl) gamma ray spectrometer due to interferences between Th and U lines and poor energy resolution of the NaI (Tl) detector. The concentration of K, U, and Th measured with NaI(Tl) for these samples are compared with those obtained from HPGe detector, shown in Table 3.8.

### Chapter - 4

# CeriumBromideGammaRaySpectrometer for Space Applications.

#### 4.1 Introduction

For many gamma-ray spectroscopy applications, a common problem is dealing with low-intensity gamma-ray emissions. This is particularly true for remote gamma-ray spectroscopy of planetary surfaces where the  $\gamma$ -ray flux is very low. In recent time, LaBr<sub>3</sub>:Ce are being the most attracting crystals among scintillators particular for the application of  $\gamma$ -spectroscopy measurements due to their good time resolution (of the order of few hundreds of picoseconds) and energy resolution (<3% at 662 keV). Additionally, the high Z<sub>eff.</sub> (Effective atomic number) of LaBr<sub>3</sub>:Ce, high density of the crystal and the availability of large volume allow the crystal for a potential use for high-energy  $\gamma$ -rays (up to 20 MeV) (R. Nicolini et. al. 2007). But due to the high intrinsic activity of LaBr<sub>3</sub>:Ce, it is difficult to estimate the concentration of K, U, and Th as discussed in chapter 3. In this chapter, the development of a gamma ray spectrometer using CeBr<sub>3</sub> has been discussed.

The recently developed Cerium doped Lanthanum bromide detector provides probably the best energy resolution as compared to the other scintillation detector and also it has an advantage that it can operate at room temperature without much compromising in light output. Thus, the Bepi-Colombo gamma ray spectrometer for the ESA/JAXA mission to Mercury (Mitrofanov et. al. 2010) uses a LaBr<sub>3</sub>:Ce detector. However, the presence of intrinsic activity due to <sup>138</sup>La and <sup>227</sup>Ac limits LaBr<sub>3</sub>:Ce's use in wide-ranging applications. The decays <sup>138</sup>La radioactive isotope degrades the detection performance, particularly for energies below 1.5 MeV. <sup>227</sup>Ac also exists as a contamination with La due to their similarity in chemical properties. The decay of <sup>227</sup>Ac produces five daughter isotopes through alpha emission which covers an energy range from 1.8 to 3 MeV.

Presently we have procured a 1" x 1" CeBr<sub>3</sub> from Scionix Holland BV. As investigated in the present study, the recently available CeBr<sub>3</sub> is an optimum compromise between an ideal <sup>138</sup>La-free- LaBr<sub>3</sub>:5%Ce and LaBr<sub>3</sub>:5%Ce itself, offering a concrete advantage over LaBr<sub>3</sub>:5%Ce for the detection of low-intensity gamma rays.

#### 4.2 Cerium Bromide Detector (CeBr<sub>3</sub>)

Both LaBr<sub>3</sub> and CeBr<sub>3</sub> crystals have the uranium tri-chloride (UCl<sub>3</sub>) lattice type with an asymmetrical hexagonal crystal structure and a non-isotropic thermal expansion coefficient which induces a tendency to crack during the cooling down following the crystal growth. The ionic radius of Ce (120 pm) is small as compared to La (122pm) (Shannon, 1976), and the Z<sub>eff.</sub> of CeBr<sub>3</sub> (45.9) is larger than that of LaBr<sub>3</sub>:5%Ce (45.3). As a result, CeBr<sub>3</sub> density is slightly larger than that of LaBr<sub>3</sub>:5%Ce. Because of the slightly larger Z<sub>eff.</sub> of CeBr<sub>3</sub>, a few percent advantage in detection efficiency compared to LaBr<sub>3</sub>:5%Ce is expected for CeBr<sub>3</sub> at energies dominated by pair production where the interaction probability rises approximately as Z<sub>eff<sup>2</sup></sub> (Quarati et.al. 2013).

The density of LaBr<sub>3</sub>:Ce is 5.07 g/cm<sup>3</sup> with melting point 783<sup>o</sup>C and the density of CeBr<sub>3</sub> is 5.18 g/cm<sup>3</sup> with melting point 732<sup>o</sup>C (Higgins et.al. 2008). The low melting points allowed us to grow the crystals in sealed quartz ampoules by using the Bridgman crystal growth technique as described earlier for crystal growth of LaBr<sub>3</sub>:Ce. Presently large sizes of CeBr<sub>3</sub> crystal are being grown by Hellma Materials GmbH and Scionix Holland BV.

#### 4.2.1 Mechanism of Scintillation of CeBr<sub>3</sub>

The scintillation mechanism of CeBr<sub>3</sub> is similar to that of LaBr<sub>3</sub>:5%Ce characterized by a Ce<sup>3+</sup> (Shah et. al. 2005, Drozdowski, 2008). The Ce<sup>3+</sup> emission is due to the transition from the lowest 5d to 4f ground state which leads to the characteristic double emission band (Quarati et. al. 2013). The peak emission wavelength for CeBr<sub>3</sub> is ~ 370 nm (Shah et. al. 2005, Quarati et. al. 2013). But for this emission spectrum shift slightly to longer wavelength with an increase in crystal thickness as shown in Figure 4.1.



Figure 4.1: X-ray emission of LaBr<sub>3</sub>:5%Ce and three different size of CeBr<sub>3</sub> (adopted from Quarati et. al. 2013).

#### 4.2.2 Characteristics of CeBr<sub>3</sub>

#### • Decay Time

CeBr<sub>3</sub> and LaBr<sub>3</sub>:5%Ce both are characterized by a short radiative lifetime resulting in an intrinsic 1/e decay time constant,  $\tau$ , of 17 ns and 15 ns, respectively (Shah et. al. 2005, Bizarri et. al., 2006). But it has been found that  $\tau_{eff.}$  increases up to ~27 ns with the increase in sample size (Quarati et. al. 2013). The rise time of CeBr<sub>3</sub> has been measured to be about 0.1 ns (Shah et. al. 2005).

#### • Light Output and Energy Resolution

Scintillation light yield for CeBr<sub>3</sub> crystal has been measured with 662 keV as 20,000 photons per MeV (Drozdowski et. al., 2008, Quarati et. al., 2013) which approximately 25% of the LaBr<sub>3</sub>:5%Ce. But the absolute light yield has been measured as > 60,000 photons per MeV (Quarati et. al., 2013, Shah et al. 2005). The light yield of CeBr<sub>3</sub> is affected by self-absorption and re-emission processes, and the corresponding larger probability of photon loss. As earlier mention LaBr<sub>3</sub>:5%Ce is best among all the scintillators which provide an energy resolution of ~3 % at 662 keV, while CeBr<sub>3</sub> provides ~4 % energy resolution at 662 keV.

#### • Intrinsic Activity

As discussed earlier, LaBr<sub>3</sub>:5%Ce has intrinsic activity due to the presence of <sup>138</sup>La radioactive and due to the presence of <sup>227</sup>Ac as contamination. Similarly, plutonium is chemically homolog to cerium and hence can induce activity like actinium in LaBr<sub>3</sub>:5%Ce. However, Billnert et. al. (2011) has shown that there is no evidence of contamination from actinium in CeBr<sub>3</sub> crystal or from plutonium. A few coincident events due to Compton scattered and cascading  $\gamma$ -rays, as well as muon-induced pulses, are also visible. In Figure 4.2 a comparison for the selfactivity from CeBr<sub>3</sub> with few other scintillation detectors are shown. It can clearly be observed that the intrinsic activity of CeBr<sub>3</sub> is lower (by an order of magnitude) compared to LaBr<sub>3</sub>: Ce and LaCl<sub>3</sub> within the energy range of 0 -3 MeV (Guss et.al, 2009).



Figure 4.2. Comparison of self-activity for 1" x 1" CeBr<sub>3</sub>, LaBr<sub>3</sub>:Ce, LaCl<sub>3</sub>, and NaI(Tl) (Adopted from Gauss et. al. 2009).

Our study will focus on the background study of CeBr<sub>3</sub> and feasibility study of CeBr<sub>3</sub> for the estimation of concentration of K, U and Th using geological samples.

#### 4.3 Feasibility Study of CeBr<sub>3</sub> for Space Application.

#### 4.3.1. Experiment Setup

The newly developed Cerium bromide (CeBr<sub>3</sub>) detector is available with different sizes starting few mm thickness to 4 inch. The characteristic details of 1" x 1" CeBr<sub>3</sub> scintillation detector as per the data sheet are listed below,



Figure 4.3. Front view of CeBr<sub>3</sub> scintillation detector.

Table 4.1: Properties of CeBr<sub>3</sub>.

Crystal	CeBr <sub>3</sub>
Size	1″ (Φ) x 1″ height
Light Yield	60,000 Ph/MeV
Decay Time	17 ns
Light output	380 nm
Energy	4 % @ 662 keV
resolution	
Operating	0 to 25º C
temperature	



Figure 4.4. Block diagram of CeBr3 gamma ray spectrometer.

The breadboard model of the CeBr<sub>3</sub> gamma ray spectrometer has been developed using commercial grade components. All the components used for the development of the breadboard model are also available in space-qualified grade and hence in future the focus will be on development of the space qualified model. The GRS design used for the developed of the gamma ray spectrometer is shown in Figure 4.4, and is mainly divided into four parts (i) Front-End-electronics (ii) Biasing Module, (iii) Data processing Unit and (iv) Data Acquisition System. The Front-End-Electronics consists of a charge sensitive pre-amplifier, shaping amplifier, discriminator, peak detector and analog-to-digital converter. A CeBr<sub>3</sub> detector  $(1'' \times$ 1") from Scionix, Holland coupled with a 2" diameter Hamamatsu PMT has been used for the present work. The pre-amplifier and voltage divider used for this measurement are also from Scionix, Holland. The required biasing voltage (-820 V) for the detector has been provided through a Canberra high voltage module 3106D. The output from the pre-amplifier is connected to the shaping amplifier module. Subsequently, the peak of the Gaussian shaped output from the shaping amplifier has been detected using a PH-300 peak detector from Ametek and then digitized using a 14-bit ADC. The digitized data are processed through FPGA and are readout in a data acquisition card through LabVIEW software. All of the above modules are developed in-house, as outlined by Panda et. al. (2017).

#### 4.3.2 Shaping Amplifier and Discriminator

The CSPA (Charge sensitive pre-amplifier) signal has been amplified using a linear amplifier. To achieve a good energy resolution, the output of the CSPA has to be amplified with an optimum peaking time. We have chosen a time constant of 2  $\mu$ s for our shaping amplifier since the change in peak position is within a few ADC channels for high count-rates (~50 kcounts/s). The bread-board model of the shaping amplifier has been designed with three stage CR-(RC)<sup>2</sup> five-pole amplification. Each stage of the shaping amplifier is designed with very low noise, high speed, wide bandwidth video amplifier IC. The 1<sup>st</sup> stage of the shaping amplifier consists of CR-high pass filter which basically acts as a differentiator. The 1st amplification stage is adjusted to a gain of 2. Both the second and third stage shaping amplifier are RC low pass filters, and the gain values for these stages are 2 and 1.2 respectively. The 3rd stage of shaping amplifier is kept in an adjustable mode to test the final shaping at different voltage level upto a maximum level of 10 Volts. With the present gain setting, the output pulse corresponds to a maximum energy of  $\sim$ 3 MeV. The gain can be adjusted to have a maximum energy range upto 10 MeV. To adjust the pole-zero of the signal, an additional resistance has been used across the capacitor, just before the 1<sup>st</sup> amplifier.

A simple discriminator circuit has been used for pulse height analysis where the discriminator is set above a noise level and produces a standard logic pulse for counting. This circuit generates a square logic pulse for each pulse of shaping amplifier and is used for event identification. The discriminator circuit is designed using a fast op-amp as a voltage comparator. The reference voltage (basically the LLD) set for the comparator acts as the minimum threshold energy that can be detected. Presently, we have fixed the threshold voltage to 80 mV, and this configuration will be made as a variable by using a DAC (8-bit).

#### 4.3.3 Peak Detector and Analog-to-Digital Converter

The Gaussian-shaped output from the shaping amplifier is coupled to a peak detector for tracking analog input pulses from the shaping amplifier and for keeping the maximum amplitude as a peak voltage on a hold capacitor during analogue to digital conversion and pulse processing. We have used AMPTEK PH300 peak detector which is available in the hybrid form with high speed, low droop rate and consumes very low power. The peak detector requires some control logic signals for peak detection which are provided through FPGA. The logic signals are so defined that the peak holds till the conversion of time of ADC and pulse processing are completed. The peak detected by the peak detector is digitized by using an analog-to-digital converter (ADC). We have used a fast 14-bit successive-approximation ADC with a 0.3  $\mu$ s Track/Hold acquisition time. The ADC is having an on-chip clock oscillator, signal conditioning circuitry, and a high-speed parallel interface. This ADC requires a reference voltage of 2.5 V which is generated by using a programmable precision voltage generator circuit. The parallel outputs from the ADC are processed by the readout system.

#### 4.3.4 FPGA Processing Electronics and Data Acquisition

In radiation spectroscopy-based applications, pulse height provides radiation energy information, whereas pulse shape of detector indicates interaction position, interaction time, type of detector material, species of interaction. Using analogue circuits, it is difficult to realize pulse rise-time measurements and can be more easily carried out by digital signal processing. The GRS readout system consists of FPGA based control logic and PCI based PC interface with LABVIEW software for data readout. The readout system development has been developed using a A3P250-PQG208FPGA manufactured by Actel. The breadboard model and the functional block diagrams are shown in Figure 4.4. The signal from the detector is amplified by the shaping amplifier and a triggered pulse is generated to track the shaping output. Once the triggered pulse is detected by the FPGA, it provides the required control signal for the peak-detector to hold the signal amplitude and initiate the analogue-to-digital conversion. The peak output is digitized by the 14-bit parallel ADC (13-bit

parallel data and 1-bit for sign). The 13-bit parallel data are read out by FPGA and then passed on to the data acquisition system.

The digitized parallel data are interfaced to computer through a PCIe-6537, National Instrumentation (NI) high-speed digital I/O interface device. Data acquisition software (DAQS) has been developed using LABVIEW to acquire the data from the processing electronics. The data readout is synchronized with the gamma ray pulse by using the reference pulse from the FPGA. This reference pulse is used as the sampling clock to initiate the data fetching from the processing electronics. The DAQS has additional features such as estimation of spectral parameters including energy resolution (Full Width Half Maximum-FWHM), peak position, and energy calibration, timer for setting the total experiment duration.

In Figure 4.4 block schematic of the CeBr<sub>3</sub> gamma ray spectrometer has been shown. The PMT required an - 820V DC biasing voltage, which has been provided through a 3106D NIM module from Canberra. The PMT output has been coupled to a preamplifier VD14-E1-X-NEG module from Scionix. The output from the preamplifier of CeBr<sub>3</sub> detector is in negative ramp type. The preamplifier output has been amplified using a 3-stage amplification and the amplified signal is digitized through the processing electronics with the help of a Actel FPGA. The details of these modules have been explained in chapter-3. All the measurements were carried out with shaping time of 2  $\mu$ s.

Gamma ray measurements were carried out using a  $1" \times 1"$  CeBr<sub>3</sub> crystal coupled to a 2" photomultiplier (PMT), voltage divider and pre-amplifier procured from Scionix. A 51 mm diameter Hamamatsu R6231-100 PMT was used for our measurements. A single breadboard model has been designed, consisting of shaping amplifier discriminator peak-detector ADC and FPGA, to improve the signal-to-noise ratio (Panda et al., 2017). The required control logic signals initiate the readout system and control peak detector and ADC in the GRS system. These logic signals were generated through the FPGA. To achieve a low background, all the measurements were carried out within a lead shield chamber of 4" thickness. The gamma ray spectrometer is designed such that the system detects every event after 5 µs from the previous interval. When a rising edge is detected by the discriminator, the peak detector starts charging its maximum by activating the gate and ramp signals, with the necessary control signals being generated by FPGA for peak detector. Once the peak detector reaches its maximum, PKDT is activated by a negative edge signal and then the Gate is deactivated so that the subsequent signals can be blocked for the next 5 µs. The voltage level of ADC data has been made compatible with FPGA by using the line driver. From the line driver, these data are passed to FPGA and then onto PCI e-digital I/O device. The digital data from the processing board were readout by data acquisition software developed using codes written using LabVIEW through a PCIe 6537-NI digital I/O device.

#### 4.3.5. Calibration

Measurement of the channel to energy conversion requires analysis of gamma ray spectra collected and fitting with Gaussian distribution functions for estimation of peak centroid and FWHM. The detector calibration has been done by using a number of radioactive sources like <sup>133</sup>Ba, <sup>137</sup>Cs, <sup>22</sup>Na, <sup>60</sup>Co, <sup>57</sup>Co, and <sup>241</sup>Am. The sources are kept at a distance of 55 mm from the detector and are measured for 600 seconds. Figure 4.5 shows the response of the CeBr<sub>3</sub> detector using various radioactive sources. The linearity curve for the CeBr<sub>3</sub> gamma ray spectrometer is shown in Figure 4.6. A good correlation has been found between the ADC channel and the energy as shown in Figure 4.6. In this Figure, the ADC channel vs energy data has been fitted with a straight line having a slope (gain) of  $\sim 0.367$  and an intercept (offset) of  $\sim$ 3.64. We have also evaluated the FWHM energy resolution at various energies for the CeBr<sub>3</sub> gamma ray spectrometer. The energy resolution at 662 and 1274 keV, measured using  $^{137}\text{Cs}$  and  $^{22}\text{Na}$  radioactive source, are  ${\sim}4.0\%$  and 2.8%respectively (Panda et. al. 2017). Quarati et al., 2013 measured the energy resolution of  $\sim 4\%$  at 662 keV for a 2"  $\times$  2" CeBr<sub>3</sub> gamma ray spectrometer. The energy resolution obtained from CeBr<sub>3</sub> were compared with that obtained from a 2" x 2" NaI(Tl) detector from Saint Gobain. Using NaI(Tl), the energy resolution obtained at 662 and 1274 keV are determined to be 6.8% and 5.2% respectively. The calculated energy

resolution from the experimental results has been compared with the theoretical energy resolution values (proportional to E<sup>-0.5</sup>) is shown in Figure 4.8.



Figure. 4.5. Response of different radioactive sources measured with CeBr<sub>3</sub> detector.



Figure 4.6: Calibration curve of CeBr<sub>3</sub>.

#### 4.3.6 Energy Resolution

The spectra obtained from the different radioactive sources are shown in Figure 4.7. All the photo-peaks have been identified and the centroid of all the photo-peak and FWHM were found out by fitting the photo-peak to Gaussian fitting using Sigma plot.









Figure 4.7: Response of radioactive sources measurement with CeBr<sub>3</sub>. (a) <sup>241</sup>Am, (b) <sup>133</sup>Ba, (c) <sup>137</sup>Cs, (d) <sup>22</sup>Na, and (e) <sup>152</sup>Eu.



Figure 4.8. Variation of energy resolution with energy for CeBr<sub>3</sub>.

The energy resolution of each photo-peak has been estimated from their photo-peak fitting. To find out the relation between the energy resolution and their corresponding energy, the energy resolution vs energy has been plotted as shown in Figure 4.8. The energy resolution estimated from the experimental data points are fitted with power law and we found that the energy resolution is related to energy as following,

 $R = 138.94 \ x \ E^{-0.55}$ 

## 4.3.7 Count Rate Variation with Change in Source to Detector Distance for CeBr<sub>3</sub>

The gamma ray instrument for planetary exploration is mainly used in orbiter mission. For example, Mars Odyssey has a science orbit  $\sim$ 400 km ((370 – 430 km)) and the gamma ray spectrometer provided GRS data with a special resolution of  $\sim$ 450 km. Similarly, the lunar prospector with a 100 km orbit provided a GRS data with the spatial resolution of 150 km and Kaguya has a polar science 100 km provided GRS data with the spatial resolution of 160 km. Therefore, the orbital distance from the planetary surfaces plays an important role. The total gamma ray received at the gamma ray detector varies with the orbital distance. In this experiment, an attempt has been made to show the behavior of the count rate with change in the source to detector distance.

In this experiment, <sup>22</sup>Na radio-active source has been used. <sup>22</sup>Na has a gamma line at 511 keV and 1274 keV. Here 511 keV gamma energy line has been taken into consideration to observe the count rate variation due to its higher count rate than the 1274 keV energy line. The experiment is carried out inside the lead shield chamber, with a wall thickness of 4 inches. The same experiment is also carried out outside the lead shield chamber. Inside the lead shield chamber the source to detector distance was varied from 1 cm to 30 cm, while outside the lead

shields the distance varied from 1 cm to 100 cm. The photo-peak counts at 511 keV were integrated and the count rate was estimated. The count rate for the different source to detector distance is given in Table-2.



Figure 4.9: Variation of photo-peak count rate expressed as a function of source to detector distance in side Pb shield of 4" wall

The count rate as a function of source to detector distance has been plotted in Figure 4.9. It has also been observed that with an increase in the source to detector distance, the energy resolution also undergoes a change. This variation of energy resolution is estimated to be within 10% (Panda et. al 2017). From this experiment, it shows that the photo-peak count rate decreases as the distance of the source is increased, as expected.

Source-Detector	Counts s <sup>-1</sup>
Distance in cm	
1	296.8
5	54.0
10	18.7
15	9.4
20	5.9
25	3.6
30	2.7

 Table 4.2: The dependence of count rate on source to detector distance.

#### 4.3.8 Response of CeBr<sub>3</sub> with Biasing Voltage

To test the ADC Channel linearity with respect to the potential voltage applied to the photomultiplier tube, we have measured the response of <sup>137</sup>Cs at different biasing voltages. The plot for the ADC Channel vs. biasing voltage is shown in Figure 4.10. It can be observed that the relation between the ADC channel with the applied potential is not purely linear. This result supports the observation of non-linear response of ADC channel with biasing voltage as suggested by Fraile et al. (2013).



Figure 4.10. Linearity for Biasing Voltage vs ADC Channel at 662 keV(<sup>137</sup>Cs).



Figure 4.11. Plot for Biasing Voltage vs. Energy Resolution at 662 keV (<sup>137</sup>Cs).



Figure. 4.12. Linearity curve for Energy vs ADC Channel at different Biasing Voltage.



Figure. 4.13. Plot for Energy resolution vs. Energy at different Biasing Voltage.

From the <sup>137</sup>Cs response curves at different biasing voltages, the resolutions are also estimated by fitting the photo-peak. The resolution obtained at different biasing voltages are plotted against the biasing voltage and is shown in Figure 4.11. From the Figure, it can be observed that the performance of the CeBr<sub>3</sub> provides a better resolution at 810 V which < 4 % @ 662 (<sup>137</sup>Cs). The correlation between the ADC channel and energy operated at different biasing voltages is shown in Figure 4.12. Similarly, in Figure 4.13 it has been shown that the energy resolution obtained at different energies with a change in biasing voltage. It can be observed from the Figure 4.11 that the encircled (in green color elliptical shape) values show the energy resolution are less than 4%. From Figure 4.11 it also can be observed the optimum biasing voltage for this CeBr<sub>3</sub> is around 810 to 820 V, where the best energy resolution can be achieved.

#### 4.3.9 Shaping Time vs Energy Resolution

The pulse width of the output signal after the amplification in shaping amplifier turned to be a function of shaping time rather than the scintillator time constant. A small shaping time constant may have better time resolution but will provide the worse energy resolution. Therefore, a proper shaping time constant should be selected to optimize and find the best energy resolution. <sup>22</sup>Na radioactive source has been used to check the variation of energy resolution for lower as well as higher energy. In this experiment, Ortech 671 shaping amplifier NIM module has been used to get a shaping time from 1 µs to 20 µs. In Figure 4.14 (a) the spectral response of <sup>22</sup>Na is shown for different shaping time constants. With the change in shaping time constant, the ADC channel also shifted as can be observed in Figure 4.15 (b) and the shift in ADC channel is not varying linearly with respect to change in shaping time. In Figure 4.15(c), it can be observed that at 511 keV, the energy resolution is well within the mean energy resolution with a shaping time from 1 to 6 µs. However, for the 1274 keV best energy resolution is in between 1 to 2 µs. Therefore, for our gamma ray spectrometer, 2 µs shaping time has been chosen.





Figure 4.14. (a) Spectral response of <sup>22</sup>Na radioactive source at different shaping time, (b) variation of ADC Channel with shaping time. (c) Energy resolution vs. shaping time.

#### 4.3.10 Intrinsic Activity of CeBr<sub>3</sub> Detector

In chapter-3, it has been mentioned that due to the high intrinsic activity it is difficult to measure the concentration of K, U, Th using LaBr<sub>3</sub>:Ce crystal (Panda at. el. 2016). Therefore, before proceeding for sample measurement, the intrinsic activity of CeBr<sub>3</sub> needs to be studied thoroughly. In order to have a lower environmental background, all the experiments have been carried out inside a 4" thick lead shield chamber. Further, the background spectrum has been measured for ~48 hours. The background spectrum from CeBr<sub>3</sub> is shown as in Figure 4.16a and Figure 4.16b the background spectrum is zoomed from 1.2 MeV to 2.2 MeV. From the background spectrum it can clearly be visualized that the total intrinsic activity of CeBr<sub>3</sub> detector is much lower than LaBr<sub>3</sub>:Ce. Figure 4.16.a. shows that there are few peaks in the energy range 1.3 MeV to 2.1 MeV. The intrinsic activity for the different energy range has been calculated as shown in Table-3.



Figure 4.15a Gamma ray background spectra measured using 1" x 1" CeBr<sub>3</sub> gamma ray spectrometer kept within a 4" thick lead shield. CeBr<sub>3</sub> detector.



Figure 4.15b. Background measurement of CeBr<sub>3</sub> in the energy range 1.3 MeV to 2.1 MeV.

Table 4.3: Intrinsic activity	v of CeBr <sub>3</sub> at different energy range.
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Detector	0.02 – 3 MeV	1.4-1.5 MeV	1.7 – 1.8 MeV	2.55-2.7 MeV
	Counts s <sup>-1</sup> cm <sup>-3</sup>			
		<sup>40</sup> K	238U	<sup>232</sup> Th
	0.11	0.000	0.000.6	0.0004
CeBr <sub>3</sub>	0.11	0.002	0.0026	0.0001
(1" x 1")				

#### 4.4 Comparison of CeBr<sub>3</sub> with LaBr<sub>3</sub>:Ce and NaI(Tl)

#### Intrinsic Activity:

The background measurements for all three detectors have been carried out inside a lead shield chamber of wall thickness 4". To have stable background, all the measurements were carried for a long integration time. The background spectra from these three detectors have been shown in Figure 4.14. The total background within the energy range of 20 keV to 3000 keV, and for the energy window of <sup>40</sup>K (1400 – 1500 keV), <sup>238</sup>U (1700 – 1800 keV) and <sup>232</sup>Th ((2550 – 2700 keV) were estimated from these spectra. The total background in the energy range 20 keV to 3 MeV for LaBr<sub>3</sub>:Ce detector was estimated to be 0.87 counts s<sup>-1</sup> cm<sup>-3</sup>, while in case of CeBr<sub>3</sub> it was nearly two order lower (0.11 counts  $s^{-1}$  cm<sup>-3</sup>). Whereas, for NaI(Tl) it was even one order less than CeBr<sub>3</sub>. In <sup>40</sup>K window the total activity for LaBr<sub>3</sub>:Ce, CeBr<sub>3</sub> and NaI(Tl) were estimated to be 0.18 counts s<sup>-1</sup> cm<sup>-3</sup>, 0.002 counts s<sup>-1</sup> cm<sup>-3</sup> and 0.0001 counts s<sup>-1</sup> cm<sup>-3</sup> respectively. For the <sup>238</sup>U window the total activity for LaBr<sub>3</sub>:Ce, CeBr<sub>3</sub> and NaI(Tl) were estimated to be 0.001 counts s<sup>-1</sup> cm<sup>-3</sup>, 0.0026 counts s<sup>-1</sup> cm<sup>-3</sup> and 0.00008 counts s<sup>-1</sup> cm<sup>-3</sup>. Similarly, in  $^{232}$ Th window the total activity for LaBr<sub>3</sub>:Ce, CeBr<sub>3</sub> and NaI(Tl) were estimated to be 0.01 counts  $s^{-1}$  cm<sup>-3</sup>, 0.0001 counts  $s^{-1}$  cm<sup>-3</sup> and 0.0006 counts  $s^{-1}$  cm<sup>-3</sup>. The activity from these detectors are listed in Table-2.



Figure 4.16. Comparison of intrinsic activities from LaBr<sub>3</sub>:Ce, CeBr<sub>3</sub> and NaI(Tl).

From Figure 4.16 it can be observed that within the 20 keV to 1.3 MeV the total intrinsic activity of LaBr<sub>3</sub>:Ce is more than two order as compared to CeBr<sub>3</sub> and NaI(Tl). But, in this range the total intrinsic activity from NaI(Tl) and CeBr<sub>3</sub> are almost same. The high intrinsic activity (0.61 counts s<sup>-1</sup> cm<sup>-3</sup>) is due to decay of <sup>138</sup>La to <sup>138</sup>Ce by  $\beta$  decay at 789 keV and due to the  $\beta$  continuum it extends up to 1050 keV. Whereas, in the energy range 1.3 to 2.1 MeV the intrinsic activity from CeBr<sub>3</sub> was estimated to be 0.014 counts s<sup>-1</sup> cm<sup>-3</sup> and for NaI(Tl) it was 0.0008 counts s<sup>-1</sup> cm<sup>-3</sup>. In the same energy range, the intrinsic activity with LaBr<sub>3</sub>:Ce was estimated to be 0.21 counts s<sup>-1</sup> cm<sup>-3</sup>. In LaBr<sub>3</sub>:Ce the high activity is mainly due to the presence of naturally radioactive <sup>138</sup>La which is decayed to produce <sup>138</sup>Ba and a 32 keV x-ray emission due to readjustment of electrons. In Figure 4.16 it can be observed with CeBr<sub>3</sub> detector there are few peaks appearing in the same energy range. These peaks purely due to alpha contamination of <sup>227</sup>Ac (Quarati et. al., 2013 and Guss et. al. 2009). In LaBr<sub>3</sub>:Ce the intrinsic activity due alpha contamination appears mainly in the energy region of 1.7 MeV to 2.7 MeV. Beyond 2.1 MeV the intrinsic activity of CeBr<sub>3</sub> is even lower than NaI(Tl).

Table 4.4 : Comparison of intrinsic activity measure with LaBr<sub>3</sub>:Ce, CeBr<sub>3</sub> & NaI(TI) at different energy windows.

Detector	0.02 – 3 MeV	1.4 - 1.5 MeV	1.7 - 1.8 MeV	2.55-2. 700 MeV
	Counts s <sup>-1</sup> cm <sup>-3</sup>	Counts s <sup>-1</sup> cm <sup>-3</sup> <sup>40</sup> K	Counts s <sup>-1</sup> cm <sup>-3</sup> <sup>238</sup> U	Counts s <sup>-1</sup> cm <sup>-3</sup> <sup>232</sup> Th
LaBr3:Ce (3" x 3")	0.87	0.18	0.001	0.01
CeBr <sub>3</sub> (1" x 1")	0.11	0.002	0.0026	0.0001
NaI(Tl) (2" x 2")	0.01	0.0001	0.00008	0.0006

#### **Energy Resolution:**

The energy resolution of CeBr<sub>3</sub> is compared with LaB<sub>3</sub>:Ce and Nal(Tl). Radioactive sources, <sup>241</sup>Am, <sup>57</sup>Co, <sup>109</sup>Cd, <sup>137</sup>Cs, <sup>133</sup>Ba, <sup>22</sup>Na, <sup>60</sup>Co, <sup>152</sup>Eu and <sup>226</sup>Ra have used to compare the energy resolution for these three detectors. The response of these sources from LaBr<sub>3</sub>:Ce and NaI(Tl) have been shown in Chapter-3. For comparison studies, the spectral responses of few sources having multienergy (like <sup>133</sup>Ba, <sup>152</sup>Eu and <sup>226</sup>Ra) from the three detector are shown in this section. From Figure 4.17 it can be observed that the spectral response from LaBr<sub>3</sub>:Ce can clearly be distinguished the photo-peak as compared to CeBr<sub>3</sub>. For Example, <sup>133</sup>Ba has four gamma energy peaks within 200 keV energy range. These peaks are well identified with LaBr<sub>3</sub>:Ce and CeBr<sub>3</sub>, but NaI(Tl) it is not shown in Figure 4.17a. Similarly, from <sup>152</sup>Eu radioactive source there are two peaks within 1050 to 1150 keV, which can be observed from the spectral response of LaBr<sub>3</sub>:Ce (Though could not be well separated), whereas only a single peak can be observed from CeBr<sub>3</sub> and NaI(Tl). This shows that LaBr<sub>3</sub>:Ce energy resolution is better that CeBr<sub>3</sub> and NaI(Tl). In Figure 4.18 the correlation between energy resolution and
the corresponding energy is shown. The variation of estimated energy resolutions with energy can be fitted using the following relations

$$R_{LaBr_3:Ce} = 103.23 \ x \ E^{-0.55}$$
$$R_{CeBr_3} = 138.94 \ x \ E^{-0.55}$$
$$R_{NaI(Tl)} = 149.92E^{-0.48}$$

A comparative study of LaBr<sub>3</sub>:Ce, NaI(Tl) and CeBr<sub>3</sub> shows that CeBr<sub>3</sub> is a better choice than LaBr<sub>3</sub>:Ce & NaI(Tl) for a gamma ray spectrometer.





Figure 4.17. Comparison of response of radioactive source from LaBr<sub>3</sub>:Ce, CeBr<sub>3</sub> and NaI(Tl). (a)<sup>133</sup>Ba and (b)<sup>152</sup>Eu.



Figure.4.18. Comparison of energy resolution vs energy correlation from LaBr<sub>3</sub>:Ce, CeBr<sub>3</sub> and NaI(Tl).

# 4.5 Estimation of K, U and Th Concentration from Geological Samples

For estimation of low-level background dominant activity, there is a need for proper measurement procedure, statistical interpretation of count data and the minimum detection limit of the instrument.

#### Minimum Detection Limit (MDL)

The minim detection limit (MDL) is defined as the minimum concentration that can be measured and reported with 99% confidence that the concentration of the sample is greater than zero and can be expressed as

$$MDL = \frac{2.71 + 4.65\sqrt{B}}{t}$$

For this CeBr<sub>3</sub> gamma ray spectrometer the minimum detection limit estimated for  ${}^{40}$ K,  ${}^{238}$ U, and  ${}^{232}$ Th were estimated to be 0.0049, 0.0048 and 0.00046 counts s<sup>-1</sup> respectively (Gilmore and Hemingway ,1995). Next attempts were made to estimate the concentration of K, U, and Th in some terrestrial samples. The gamma ray spectrum for some potassium salts (KCl and K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub>) was also measured to check the signal-to-noise ratio, as shown in Figure 4.19.







Figure 4.19. Comparison of background spectra and K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub> and gamma ray spectra from KCl measured with CeBr<sub>3</sub> detector.

From the Figure 4.18 and 4.19, it can be observed that from the spectra of both salts, the K peaks at 1461 keV can be easily identified.

For the estimation of K, U and Th concentration, granite rock has been powdered using Mini Ball to a grain size of ~200  $\mu$ m. The granite rocks were collected from Godhra, Gujarat granitic belt. In this experiment GHRF#2, a soil sample measured has been used as the standard. The concentration K, U, and Th of GHRF#2 was estimated using HPGe gamma ray spectrometer where STD#107 (K - 2.6%, U - 5.7 ppm, and Th - 14.5) used as the standard. Using HPGe gamma ray spectrometer, the concentration of GHRF#2 for K, U, and Th were estimated to be 3.38%, 14.7 ppm and 48.7 ppm respectively. The gamma ray spectra for the standard and the sample were integrated for 6 hrs. At the first attempt, unprocessed (raw) data from the CeBr<sub>3</sub> gamma ray spectrometer were used and the K concentration of sample 3A was estimated to be 4.2%, which was ~13% higher than the HPGe based value of ~3.7%. Then the gamma ray spectra were smoothen using running average method. Using the smoothed spectra K estimation was improved, and the K concentration of sample 3A was determined to be 3.8%. The concentration of U in sample 3A was estimated using the 1764 keV ( $^{214}$ Bi) gamma line. Using processed spectra, we could estimate the concentration of U to be 2.1 ppm, which agrees very well with an HPGe based U concentration of 2.04 ppm. Estimation of Th using the CeBr<sub>3</sub> could not be performed using 2615 keV line due to the low efficiency of the 1" x 1" detector at this energy (Panda et al. 2017). Other low energy gamma lines from Th could also not be utilized due to low energy resolution, and since there are interferences due to gamma lines from other elements (e.g., U). The Th concentration could not be estimated due to the poor efficiency of the 1" x 1" detector. With a large detector (e.g. 2" x 3") detector estimation of Th is likely to be possible.

# Chapter - 5

# Methodological Investigations of Infrared Stimulated Luminescence Signals from Feldspars.

#### 5.1 Introduction

The suggestion of Daniel et. al., 1953 that thermoluminescence (TL) could be used for geological and archaeological age evaluation provided a stimulus for basic research on methodology and application of TL dating (Aitken et. al., 1964). The method was fully established as a reliable method for dating pottery and was also extended to sediments baked by the lava flow towards the year 1970s. Huntley, Johnson and Wintle's studies on ocean sediment cores provided evidence for an increase in TL signal with depth which suggested resetting of the TL signal occurs due to sunlight exposure for unheated sediments. This was followed by the development of various TL techniques (Wintle and Huntley, 1982; Singhvi et. al. 1982; Proszynska, 1983; Debenham and Walton, 1983) and their application to date various types of deposits, e.g., loess, marine sediments, dune sands, soils, lake sediments, fluvial sands, glacial dust, colluvium etc. With the advent of optically stimulated luminescence (OSL) dating and infra-red stimulated luminescence (IRSL) dating techniques in the 1980s, reliable dating of poorly bleached sediments (sediments having received a short pre-depositional sun-exposure) is now achievable. The luminescence methods have the potential to revolutionize Quaternary Geochronology as they can be used for dating a variety of earth surface

processes and sediments that were conventionally considered undatable and have a dating range of  $1 - \geq 200$  ka which is complementary to the radiocarbon, U-series and paleomagnetic dating methods. Secondly, the luminescence methods involve analysis of minerals constituting the sediments, thereby circumventing problems associated with sample-strata correlations. Finally, luminescence ages do not require calibration as is necessary in radiocarbon dating. As conventional radiometric methods have often been found to be inapplicable and occasionally work explored the unreliable. the present possibility of utilizing thermoluminescence and infra-red stimulated luminescence dating techniques.

#### 5.2 Luminescence

The emission of light (excluding black body radiation) from a substance subsequent to its excitation by ionizing radiation is called luminescence. The stimulated luminescence process can be explained in terms of the band theory of solids as shown in Figure 5.1.



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.

Exposure to ionizing radiations creates many free electrons and holes in the crystal. Most of these free charges recombine instantaneously. A small fraction of

these charges get trapped at lattice defect sites (viz., vacancies, interstitial and impurity atoms etc.) within the crystal. Thus, e.g., a negative ion vacancy acts as an electron trap since there exists a local deficiency of negative charge. The electron/hole traps are represented as localized energy levels between the conduction and valence bands. Eventually, the trapped charges (electrons/holes) may absorb thermal/optical energy and be released into the conduction/valence band. Some of these charges may recombine at a luminescence center to emit light. Different kinds of luminescence process exist depending upon the source of stimulation viz., thermoluminescence (heat energy), optically stimulated luminescence (visible light energy) and infra-red stimulated luminescence (infrared light energy).

#### **5.3 Thermoluminescence Dating.**

Luminescence (TL/IRSL/OSL) can only be used for dating particular events which reset the acquired luminescence of the luminescent minerals comprising the sample to zero or a negligibly low value. After resetting the event, exposure to natural radioactivity ( $\alpha$ ,  $\beta$  and  $\gamma$  radiation primarily from <sup>238</sup>U series, <sup>232</sup>Th series, <sup>40</sup>K and cosmic rays) builds up the thermoluminescence level of minerals constituting the sample. On excavation, the thermoluminescence level of a sample is a measure of the radiation dose absorbed by the minerals since the resetting/zeroing event and can be expressed in terms of the annual dose received by the constituent minerals of the sample and the time since the zeroing event. The thermoluminescence age can be expressed as (Aitken, 1985).

$$A = \frac{Paleo \ Dose}{Dose \ Rate}$$
$$= \frac{P}{aD_{\alpha} + D_{\beta} + D_{\gamma} + D_{c}}$$

Where, a is the alpha-efficiency factor , P stands for paleo dose,  $D_{\alpha}$ ,  $D_{\beta}$ ,  $D_{\gamma}$  and  $D_c$  are annual dose components for alpha, beta, gamma and cosmic respectively.

#### **5.3.1 Annual Dose Determination**

The annual dose components can be estimated by measuring the concentration of potassium, uranium, and thorium (K, U, and Th) in the sample. These elemental concentrations are used along with the "infinite matrix" assumption to evaluate the annual dose. In the "infinite matrix" approach, it is assumed that in a volume with dimensions greater than the range of the  $\alpha$ ,  $\beta$ ,  $\gamma$ radiation and cosmic rays, conservation of energy will imply that the rate of energy per unit mass is equal to the rate of energy emitted per unit mass provided that radioactivity per unit mass and absorption coefficient per unit mass is constant over the sample volume. The evaluation of annual dose by "infinite matrix" approach and the TL age equation assume that the radionuclides <sup>238</sup>U, <sup>232</sup>Th and their daughters are in equilibrium and no change in K concentration occurs since the 'zeroing event', thus ensuring that the annual dose remains constant. However, disequilibrium in the <sup>238</sup>U, and <sup>232</sup>Th series can occur in certain situations. U, Th, Ra can be leached out by percolating groundwater, especially in coarse grain deposits due to greater permeability compared to finegrained deposits (Berger, 1988).

#### Measurement Techniques

Various technique exists for determination of the annual dose components. In this thesis work the U, Th and K concentration were determined with gamma ray spectrometer using 87 mm diameter High Purity Germanium (HPGe). The detector is coupled to a pre-amplifier and a linear amplifier. The output from the amplifier is then digitized and interfaced to computer through a multi-channel analyser. The samples were crushed to grain sizes of  $\sim 200 \ \mu\text{m}$ . using mini planetary ball Mill and packed in a sealed plastic box for one month to attend the radioactive equilibrium. For the estimation of the concentration of Th, U and Th very small energy windows are chosen around 583 keV, 609 keV and 1.46 MeV for Th, U and K respectively. The cosmic-ray dose was assumed to be 150  $\mu$ Gy/a (Prescott and Stephan, 1982).

#### 5.4 Infra-Red Stimulated Luminescence Dating

In the thermoluminescence dating of sediments, a major difficulty arises in the evaluation of the I<sub>0</sub> and therefore in estimating Q. This occurs because heating the samples releases electrons from both the light-sensitive traps and the lightinsensitive traps. In nature, bleaching due to sunlight only evicts electrons from the light sensitive traps. This can be replicated in the laboratory if the light energy is utilized for stimulating the sample. In a classic paper Huntley et. al. (1985) reported on the application of monochromatic green laser light to reset a rapidly bleachable optically stimulated luminescence. An argon ion laser was used for excitation of the sample. The PMT was heavily filtered to differentiate between the excitation and emission wavelengths (blue/violet). In an attempt at dating sedimentary deposits using OSL, Huntley et. al. (1985) found that the luminescence signal increased with depth in two geological sequences spanning 0-700 ka (South-east Australian beach dunes) and 0-6 ka (British Columbian soil horizons). The additive dose method was used to evaluate the paleodose. As mentioned above, the stimulation signal could rapidly bleach the luminescence signal.

The OSL dating technique is advantageous to its TL counterpart in the following ways: (i) a negligibly low post-bleaching residual signal ( $I_0 \sim 0$ ) which makes it feasible to date poorly bleached sediments, (ii) no requirements of heating means that experiments can be performed rapidly and simply and with improvement in S/N ratio, (iii) avoidance of spurious TL and thermal quenching processes and accessibility to electron residing in deep traps. Also, since the method involves no heating there exists a possibility of dating minerals (e.g. organic compounds, gypsum) which undergo a phase change on heating.

In 1988, Hutt et.al., reported the presence of a strong resonance in the stimulation spectrum of K-feldspars in the region 750-1000 nm. This infra-red stimulated luminescence (IRSL) was eventually used for dating sediments. In comparison to Green Light Stimulation Luminescence (GLSL), IRSL is better suited for dating because of: (i) availability of a large wavelength band for detection resulting

enhanced sensitivity; (ii) faster bleaching, so a lower  $I_0$  and (iii) it's cheaper costs (Aitken, 1984). Hutt et.al. 1988 have proposed a model to explain the IRSL mechanism (Figure 5.2.): electrons are raised to excited states under IR stimulation and then arrive at the conduction band on absorbing thermal energy from lattice vibrations. Some of them recombine with holes at recombination centers to emit infra-red stimulated luminescence. IRSL is thus strongly depended upon temperature.



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

In IRSL dating, the paleodose is usually determined by the additive method or the regeneration method. Aitken and Xie (1992) recommended the use of the subtraction technique where the value of IRSL intensity after a long time interval (~200 s) is subtracted from all IRSL intensities for an aliquot. This eliminates signals from scattered IR, PMT dark count and any 'hard-to-bleach' component which remained unbleached on deposition. The light intensity of short IR exposure (short-shine) of 0.5 or 1 second is used to normalize the IRSL intensities (natural normalization) obtained on exposure for a longer time interval (shine-down  $\sim$ 12 s). Shine –down curves plots of IRSL intensity with diode exposure time) for aliquots with different amounts of artificial irradiation are constructed. Growth curves (plots of IRSL intensity with irradiation dose at a particular exposure time) are then utilized for calculating paleodose at each shine time. The paleodoses are then plotted against shine time and a shine-down plateau is a good indication of efficient zeroing of the IRSL at deposition (Aitken, 1994).

#### **Error Estimation**:

Estimation of error in luminescence ages in this thesis was based on Aitken (1985). The total error in TL age consist of two parts: (i) random error in estimation of paleodose/equivalent dose, a value, supralinearity intercept and annual dose and (ii) systematic errors arising due to uncertainty in  $\beta$  source, and K, U and Th analyser calibrations, parameter used in converting U and Th concentration to annual dose components, degree of radon escape and uncertainty in the water content. The random errors in paleodose/equivalent dose mostly arise from curve fitting. Counting statistics (determination U, Th and K) contribute to random errors in annual dose.

# 5.5 Infrared Stimulated Luminescence Signals from Feldspars

Determination of luminescence ages of feldspar grains has been the research focus of several recent studies, particularly since the luminescence signal from feldspar (Aitken, 1985; Auclair and Lamothe, 2003; Huntley and Lamothe, 2001) has potential for measuring large doses of the order of thousands of Grays. However, measurement of equivalent doses in feldspar extracts using a TL/OSL signal which originates from electron traps that are stable over geological timescales and does not exhibit anomalous fading has generally proven to be very difficult. To this end, it has been recently shown that the use of higher stimulation temperatures (225<sup>o</sup> C) in optically stimulated luminescence dating results in

lower laboratory fading rates (Thomsen et al. 2008; Buylaert et al. 2009; Murray et al. 2009), as a consequence of decreasing tunnelling probability. A post-IR IRSL signal based Single Aliquot Regeneration (SAR) procedure is being increasingly utilized for feldspar dating, after the first study of Buylaert et al. (2009) where the fading correction factor was shown to be reduced by more than 10% while using post-IR IR signals for equivalent dose measurements. While these methodological developments are crucial for Quaternary geochronology, it is equally important to perform basic studies to investigate the source and the stability of the IRSL signal being used in feldspar dating.

Duller and Bøtter-Jensen (1993) measured the ratio of IRSL signals from  $((Natural+\beta)/Natural)$  irradiated aliquots as a function of preheat temperature, and observed a nearly constant value with increasing preheat temperatures. In another study (Duller 1994), pulse annealing curves showed the presence of thermally unstable IRSL signals in laboratory irradiated aliquots (Natural+ $\beta$ ) and thus a preheat of 220° C for 10 minutes was proposed in equivalent dose measurement procedures. Although several investigations have previously shown that the infrared stimulated luminescence signal from feldspar does not arise from a specific region of the TL curve, Murray et al. (2009) observed that low temperatures TL peaks in their samples did not significantly contribute to IRSL in their samples. We investigate the source and thermal stability of the IRSL signal in two feldspar samples obtained from NIST, viz., Standard Reference Material (SRM) 99b (soda feldspar) and K-feldspar SRM 607. We have investigated the dependence of equivalent dose on preheat temperature on feldspars extracts collected from Sutlej river terrace gravels (SUN-1-GR and SHRD-1; Sharma et al. 2016).

#### 5.6 Samples

Two feldspar samples used in this study were procured from NIST (National Institute of Standards and Technology), and included a soda feldspar SRM 99b (SRM = Standard Reference Material) and a K-feldspar sample SRM 607. The Na concentration in SRM 99b is ~5%, whereas the K concentration in SRM

607 is  $\sim$ 9 wt.%. Two additional samples (SUN-1-GR and SHRD-1) were collected from a sand lens within Sutlej river terrace gravels.

## 5.7 Sample Preparation Methodology

- a) The 63-150  $\mu m$  grain size fractionation was done through wet sieving.
- b) The fractionated sample is sequentially treated with 1 N HCl solution for complete removal of carbonate from the sample.
- c) After a thorough wash with double distilled (DD) water and decantation, the sample is given 2-3 treatments of 30 % H<sub>2</sub>O<sub>2</sub> so as to remove the organic impurities. The sample is again washed with DD water, alcohol and after decantation, oven dried at 40-50° C.
- d) The light mineral fraction is used for separation of quartz and feldspar grains using a freshly made solution of Na poly-tungstate with an accurate specific gravity of 2.58 g/cm<sup>3</sup>. The separating flasks containing quartzfeldspar are left undisturbed for 10-12 hrs. Once the quartz grains are settled at the bottom of flasks, these are carefully filtered out. The feldspar grains are also filtered out and preserved after a thorough washing with DD water and alcohol, dried and packed in opaque packets.
- e) The quartz crop is treated with 45% HF in Tephlon beakers for 40 minutes (with intermittent stirring) to etch out the 20-micron outer layer of grains. After washing with alcohol, the quartz grains are treated with concentrated HCl for 20 minutes. These are washed 5-10 times with DD water and alcohol and dried at 50-60° C in the ovens. The dried quartz grains packed in small bottles in the opaque packets.
- f) About 100-200 feldspar grains are temporarily mounted on 9.8 mm diameter stainless steel discs with the help of silicon spray. Initially, about 20 such discs are prepared of each sample for pre-dating tests and luminescence measurements on the TL/OSL Reader. The discs are gently placed in the disc holes of the sample holder of TL/OSL Reader.

#### 5.8 Experimental Procedures and Samples

An automated Risø TL/OSL reader (shown in Figure 5.3.) (Bøtter-Jensen et al., 2003) was used for all TL and IRSL measurements. Photon counting used a Thorn-EMI 9235 QA photomultiplier tube coupled to Corning 7-59 and Schott BG-39 detection filters. IRSL stimulation was performed using 875 nm light emitting diodes providing between 100-200 mW cm-2 at the sample position. A calibrated <sup>90</sup>Sr/<sup>90</sup>Y beta source was used for all laboratory irradiations in our experiments.



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

## 5.9 Thermoluminescence Glow Curves

Our first experiment involved investigation of the shape of the TL glow curve. Aliquots of both samples (SRM 607 and SRM 99b) were heated to 500°C to

remove any natural thermoluminescence signals and were then given a dose of ~1.8 Gy. The aliquot was then heated to  $500^{\circ}$  C while its TL signal was measured. Figures 5.4a and 5.4b represent the plots of TL against temperature for the SRM 607 and SRM 99b aliquots. Another aliquot was treated in a similar manner but with the following difference; before measuring its TL signal, the aliquot was preheated at 250° C for 60 s. For SRM 607, the TL glow curve consists of a several peaks at ~95° C, 160° C, 260° C and 340° C. The preheated aliquot for SRM 607 shows an increase in TL intensity at temperatures beyond 250° C, and a thermoluminescence peak at about 340° C. The TL intensity remains above background levels at temperatures in excess of 400° C, and a small second TL peak at ~390° C can be inferred from glow curve shape in the K-feldspar sample (SRM 607).





Figure 5.4 TL glow curves for SRM 607, SRM 99b and SHRD-1 obtained using a heating rate of 2<sup>o</sup> C s<sup>-1</sup>.

In the soda feldspar SRM 99b, a broad TL peak is observed at temperatures between  $140^{\circ}$  -190° C; this TL signal is absent in the preheated SRM 99b aliquot, and a TL peak at ~300° C is observed in the glow curve. However, in this sample, the high-temperature TL peak cannot be observed from the TL glow curve. Murray et al. (2009) observed TL peaks at ~310° C and ~400° C in natural TL glow curves of K-feldspar extracts, and an additional TL peak between ~100°-130° C in the regenerated TL signals from their samples.

#### **5.10 Pulse Annealing Experiments**

One of the primary assumptions in the determination of equivalent dose (the laboratory dose needed to regenerate a luminescence level equal to the natural luminescence level of a sample) is that luminescence signals are stable over geological timescales. Thus, it is necessary to devise a preheating procedure which isolates a thermally stable OSL signal for estimation of equivalent dose, De. Our next experiment investigated the effect of preheating on regenerated IRSL signals from SRM 607 and SRM 99b. An aliquot was heated to 500<sup>o</sup> C for complete resetting of the IRSL signal, before being given a dose of ~9 Gy. The aliquot was then preheated at 80° C for 10 s, and its regeneration IRSL signal was measured (Li). These measurements were repeated once to ensure no phototransfer/thermal transfer effects, and a third measurement cycle was performed with a fixed preheat temperature of 80° C (T<sub>i</sub>). This measurement cycle was repeated a further 18 times with preheat temperatures being increased in steps of 20° C between 100° C and 440° C. The normalized IRSL signals shown in Figure 5.5 is from L<sub>i</sub>/T<sub>i</sub> estimates, normalized to their initial value.



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.

The regenerative IRSL pulse anneal curves show loss of IRSL signal due to thermal erosion at temperatures beyond 200° C and 150° C for SRM 99b and SRM 607 respectively, and complete thermal erosion of IRSL signals beyond 400° C. In the K-feldspar SRM 607, the IRSL signal is reduced to ~70% of its initial value at 200° C. In contrast, less than 1% of the IRSL signal is lost at 200° C for the sodafeldspar SRM 99b. Murray et al. (2009) observed that the natural TL signal became significant at temperatures higher than 200° C, which seemingly correlates with

the loss of IRSL signal at these temperatures. However, as outlined in Murray et al. (2009), and for the samples in our study, most of the loss in IRSL occurs at temperatures where the TL signal detected is much lower in comparison to TL signals below 200<sup>o</sup> C.

# 5.10.1 IR Stimulation and TL Measurement without Preheating

To investigate the relationship between IRSL and TL, we have examined the effect of infrared stimulation on the TL signals in the soda and K-feldspar samples (SRM 99b and SRM 607). An aliquot from each sample was heated to 500° C and was then given a dose of ~9 Gy. The aliquots were heated to 500° C to measure the regeneration signal. The same aliquots were then given a second regeneration dose of ~9 Gy, followed by 10 second infrared stimulation at 60° C. The regenerated TL signal was measured by heating to 500° C. This measurement cycle was repeated twice more on the same aliquot, with infrared stimulation exposures of 100 and 1000 seconds prior to measurement of regeneration IRSL signals.

In the K-feldspar sample (SRM 607), although IR stimulation reduces TL signals throughout the entire glow curve (Figure 5.6), the most significant loss of TL due to IR stimulation occurs at the 95° C TL peak. The loss of TL after 1000 s of infrared stimulation is around 98% at the peak position of ~95° C, while it is approximately 40% at the temperature of  $350^{\circ}$  C. For SRM 99b, the loss of TL signal occurs mostly between the temperature range of  $100^{\circ}$  –  $300^{\circ}$  C. The loss of TL after 1000 s of infrared stimulation is ~95% at ~95° C, while it is approximately 75% at 350° C. In an experiment to study the effect of prior infrared exposure on the TL signals from K-feldspars, Murray et al. (2009) observed large losses of TL signal at the ~140° C and ~400° C peaks. The lost TL peak positions remained at the same temperature after prior IR stimulation. These phenomena can be explained as proposed earlier by Duller (1995) involving the presence of two types of IRSL traps, one which participates in TL production, and the other trap being the primary source of the IRSL signal, but does not generate any

138

significant TL signal. Murray et al. (2009) proposed an alternative explanation wherein the TL recombination probability is decreased by the effect of infrared stimulation, affecting the recombination of charge originating from both traps in a similar proportion.



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

# 5.10.2 Effect of IR Stimulation on TL Signals Following Preheating at Elevated Temperatures.

The next experiment investigated the effect of infrared stimulation on TL signals for the feldspar samples following high temperature preheat treatments. The TL signals lost from IR stimulation are shown for two different preheat temperatures, viz., 250° C and 320° C (1 minute), in Figures 5.7 and 5.8. Both sets of data show the presence of a single TL peak centered around 300° C and 310° C for SRM 99b and SRM 607 respectively. The photo-transferred TL (PTTL) signals are negligible for both the soda-feldspar and K-feldspar. samples after preheating at 250° C followed by infrared stimulation. When this measurement cycle was repeated with a 320° C preheat temperature, photo-transferred TL peaks are visible at 120° C and 250° C in SRM 607 due to charge transfer. PTTL peaks are

also observed in TL glow curve of SRM 99b, but the relative intensities are much lower in comparison to SRM 607. Murray et al. (2009) argued that the lost TL peak observed at 410° C remained largely unaffected by IR stimulation time, and thus the IRSL signal originated primarily from TL traps above 320° C. However, in both the samples SRM 99b and SRM 607, the only significant loss of TL during IR stimulation occurs at around 300° C.



Figure 5.7a: TL lost due to IR exposure for SRM 607 measured after preheating at 250<sup>o</sup> 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<sup>o</sup> C for 60 seconds and inferred

#### 5.11 Discussion

Murray et al. (2009) investigated the variation of IRSL equivalent dose with preheat temperature (preheat plateaus) for the K-feldspar extracts considered in their study. As no systematic variation in equivalent dose could be observed with preheat temperature, the authors concluded that natural IRSL signals do not originate significantly from low temperature traps. We have used a SAR procedure (Murray and Wintle, 2000) with the same thermal treatment given to natural, regenerative and test dose aliquots extracted from the two feldspar sediments (SUN-1-GR and SHRD-1) in this study. Fading rates (*g*-value) were measured for two preheat temperatures 120° C and 320° C. The *g*-values for SUN-1-GR were 5.1  $\pm$  0.4 %/decade, and 6.7  $\pm$  1.5 %/decade (t<sub>c</sub> = 2 days) for 120° C and 320° C preheat temperatures respectively, and the values are similar within error limits. The *g*-values for SHRD-1 were 3.3  $\pm$  0.6%/decade, and 7.6  $\pm$  2.2%/decade (t<sub>c</sub> = 2 days) for the 120° C and 320° C preheat temperatures respectively. U, Th and K concentrations for these samples were measured using an HPGe gamma ray spectrometer shown in Table 5.1. (Sharma et al., 2016). Assuming a water content

of 10%, and an alpha efficiency factor of 0.1, the dose-rates for SHRD-1 and SUN-1-GR are estimated to be ~3.86 and ~5.2 mGy a<sup>-1</sup>. Table 5.2. and 5.3 show the doserate estimated from quartz and feldspar for SHRD-1 and SUN-1 samples. It is interesting to note that the fading-uncorrected IRSL age for SHRD-1 is ~19 ka, and is more than twice than the quartz age (8.5 ± 0.7 ka) obtained using a modified SAR procedure from this sample (Sharma et al., 2016). However, the quartz extracts exhibited large IR intensities (Sharma, personal communication) after laboratory irradiation; hence further work, including a fading test on the quartz extracts used in the previous study is necessary to confirm the validity of the existing ages.





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

Table 5.1: U, Th and K concentration measured using Gamma Ray Spectrometer.

Sample	U	Th	K	Cosmic Ray
	ррт	ppm	%	μGy/a
SHRD-1	$3.65\pm0.05$	$\begin{array}{c} 14.03 \pm \\ 0.25 \end{array}$	$2.15\pm0.02$	221±66.3
SUN1-GR	$7.32\pm0.17$	$26.6\pm0.9$	$1.66\pm003$	222±66

Table 5.2: OSL age measured using quartz mineral.

Sample	De (Gy)	Dose rate	Age (ka)
SHRD-1	32±1.5	3.8±0.5	8.5±0.7
SUN1-GR	27±1.1	5±0.3	5.5±0.4

Sample	De (Gy)	Dose rate	Age (ka)
SHRD-1	32±1.5	~3.86	~ 19
SUN1-GR	27±1.1	~5.2	-

Table 5.3: IRSL age measured using feldspar mineral.

Figure 5.9, suggests that for both feldspar samples considered here, 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). From the preheat plateaus presented in this paper, we agree with the above conclusion, and provide further evidence that low temperature TL peaks do not make a significant contribution to IRSL in our feldspar samples. This inference renews the debate regarding the necessity for preheating in dose estimation procedures for feldspar samples.

# **Chapter-6**

# **Conclusion and Future Scope.**

#### 6.1 Summary of Results:

This thesis presents the development of gamma ray spectrometers using scintillation detectors, viz., LaBr<sub>3</sub>:Ce and CeBr<sub>3</sub>. Attempts were made to estimate the concentration of K, U, and Th in terrestrial samples using both these two detectors, in light of our objective being determination of the surface composition of planets.

# 6.1.1 Development of a Cerium doped Lanthanum Bromide Gamma Ray Spectrometer for Planetary Missions and Feasibility Studies

High energy (>100 keV) gamma ray spectroscopy is an important technique for remote sensing studies of the chemical composition of planetary surfaces and has been used to study the surface composition of the Moon, Mars, and Asteroids at various spatial resolutions. We have developed LaBr<sub>3</sub>:Ce gamma ray spectrometer for a future planetary orbiter mission with the primary objective of determining the abundance and distribution of Th, U, K, and other major elements including Fe 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. Additionally, we have investigated the possibility of using a LaBr<sub>3</sub>:Ce gamma ray spectrometer for determination of U, Th, and K in dose-rate estimation for luminescence dating (Aitken, 1985, Murray et al. 1987). The energy resolution of the LaBr<sub>3</sub>:Ce gamma ray spectrometer using front-end and processing electronics developed in-house has been measured at 511 and 1274 keV to be 4.1% and 2.5% respectively. Our results suggest that the intrinsic activity count-rate for our  $3'' \times 3''$  LaBr<sub>3</sub>:Ce gamma ray spectrometer is ~61 counts s<sup>-1</sup> (i.e. ~0.18 counts s<sup>-1</sup> cm<sup>-3</sup>) for the  $^{40}$ K energy window (1400-1520 keV) and  $\sim$ 3.4 counts s<sup>-1</sup> for the <sup>232</sup>Th (2550-2700 keV) energy window. Although this large intrinsic activity of the LaBr<sub>3</sub>:Ce crystal inhibits estimation of the concentrations of Th and K, our attempts using a NaI (Tl) gamma ray spectrometer (with electronics developed in-house) were more successful. The Th concentration of US#110 was estimated to be  $\sim$ 11.4 ppm and is within 14% of the 13.2 ppm value determined using an HPGe gamma ray spectrometer. The K concentration of US#110 was estimated to be 0.87% and is within  $\sim 10\%$  of the 0.78% value determined independently using an HPGe gamma ray spectrometer. U estimation was not possible using the NaI (Tl) gamma ray spectrometer due to interferences between Th and U lines and poor energy resolution of the NaI (Tl) detector.

# 6.1.2 Cerium Bromide Gamma Ray Spectrometer for Space Applications

The development of a CeBr<sub>3</sub> gamma ray spectrometer has been presented here with the primary objective of determining the abundance and distribution of Th, U, K, Fe, Al and Si 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. The energy resolution of the CeBr<sub>3</sub> 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″ x 1″ CeBr<sub>3</sub> gamma ray spectrometer is ~0.03 counts s<sup>-1</sup> for the <sup>40</sup>K energy window (1400-1520 keV), and ~0.001 counts s<sup>-1</sup> for the <sup>232</sup>Th (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 an HPGe gamma ray spectrometer. The K concentration of sample 3A was estimated to be 3.8% and is consistent with the 3.7% value determined independently using an HPGe gamma ray spectrometer.

#### 6.1.3 Methodological Investigations of Infrared Stimulated Luminescence Signals from Feldspars.

Determination of luminescence ages of feldspar grains has been the research focus of several recent studies, particularly since the luminescence signal from feldspar has the potential for measuring large doses (~kGy). Reflectance spectra of quartz samples show absorption features at  $\sim$ 1.4 µm, 1.9 µm, 2.2 µm and 2.3 µm. The Na feldspar sample indicated absorption at 1.9 and 2.2 µm, whereas the K-feldspar sample absorption features occur at 1.4, 1.9 and 2.2 µm. These absorption features are associated with OH and SiOH. The reflectivity values associated with feldspar samples are observed to be at least twice in comparison to quartz samples. Further, 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 ~95° C, 160° C, 260° C and 340° C. In the soda feldspar SRM 99b, a broad TL peak is observed at temperatures between 140<sup>o</sup>-190<sup>o</sup> C; this TL signal is absent in the preheated SRM 99b aliquot, and a TL peak at  $\sim$  300<sup>o</sup>C is observed in the glow curve. Although IRSL signals from the soda-feldspar SRM 99b indicate no significant depletion at temperatures below 200<sup>o</sup> C, the pulse anneal data for the K-feldspar sample SRM 607 does show a 30% loss in IRSL while heating upto 200<sup>o</sup> C. For both natural feldspar extracts considered here (SUN-1-GR and SHRD-1), the equivalent dose value remains constant for preheating temperatures between 80° C – 320° C. It has been previously argued that a single trap (around 410<sup>o</sup> C) may be primarily responsible for the IRSL signal (Murray et al. 2009). From the preheat plateaus presented in this paper, we agree with the inference that lowtemperature TL peaks do not make a significant contribution to IRSL in several feldspar samples.

In summary, the thesis work pursued new directions to the application of new scintillation based gamma ray spectrometers in planetary sciences. Further, our work focused on luminescence techniques in earth sciences. This study contributes significantly towards methodological establishment of the applications of luminescence dating techniques and will provide a basis for routine application of the methods. Future research should focus on production and application of CeBr<sub>3</sub> crystals with ultra-low background in the development of gamma ray spectrometers. Additionally, development of an HPGe detectors with miniaturized cryocoolers (~0.5 kg) should be pursued in future laboratory and space-borne experimental studies.

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