PROGRESS ON THE DEVELOPMENT AND TESTING OF NEW SCINTILLATORS FOR PLANTARY GAMMA-RAY SPECTROSCOPY. T. H. Prettyman[1], A. Burger[2], N. Yamashita[1], R. Hawrami[2], E. Ariesanti[2], E. Rowe[2], V. Buliga[2], N. Pearson[1], M. E. Landis[1], [1]Planetary Science Institute, Tucson AZ (prettyman@psi.edu), [2]Fisk University, Nashville TN.

Introduction: Since the 1960s, planetary gamma ray emissions have been measured by sensors deployed on orbiting spacecraft and landers [1]. Gamma rays are produced by the decay of natural radioelements found in rock-forming minerals and by the steady interaction of cosmic rays with surface materials. The gamma ray leakage spectrum can be analyzed to determine the elemental composition of the regolith to depths up to a meter, providing constraints on planetary evolution and formation, and complementing information provided by other remote sensing methods.

The earliest planetary spectrometers were scintillators, which are transparent to ultraviolet and visible light emitted in response to excitation by ionizing radiation. For example, the interaction of gamma rays with the scintillator produces swift elections that ionize the medium. Scintillation light is detected by an optical sensor, such as photomultiplier or photodiode and converted to a pulse height spectrum. Peaks in the spectrum identify specific nuclear reactions occurring within the planetary surface, from which the target element/isotope can be identified (Fig. 1). The intensity of the peaks is proportional to elemental concentration, subject to correction for various systematic effects.

The energy resolution currently achieved by scintillators is inferior to cryogenically-cooled, intrinsic germanium, the "gold standard" for gamma-ray spectroscopy [e.g. 2]. Nevertheless, scintillators have ample resolution for many applications and they fill an important niche for missions that require rugged, low-cost sensors that operate at ambient temperatures, with minimal operational complexity and low impact on payload resources. We describe the development, testing and evaluation of the next generation of scintillators, including europium-activated strontium iodide, SrI_2:Eu [e.g. 3], and self-activated Cs_2HfCl_6 (CHC) [e.g. 4]. These scintillators have attributes attractive for planetary applications, including improved energy resolution compared to scintillators with flight heritage, such as bismuth germanate (BGO) [5], and high efficiency for gamma ray detection. They are not hygroscopic and are easier to grow due to their cubic structure.

Scintillator selection: Gamma ray energy resolution is fundamentally controlled by the light yield of the scintillator, the number of photons produced per MeV of energy deposited by swift electrons. Statistical fluctuations in the number of photons detected results in broadening of the gamma ray peaks. Moreover, all scintillators exhibit nonproportionality between light yield and electron energy for energies lower than about 100 keV. This results in additional variability in light production, further broadening gamma ray peaks. Nonproportionality is an intrinsic property of scintillating materials and is a major factor limiting energy resolution [6, 7]. For example, despite having lower light yield, CLYC has better energy resolution than NaI(Tl), (Table 1). For CLYC, contributions from nonproportionality are relatively low compared to NaI(Tl). Improvements in the resolution of inorganic scintillators have resulted primarily from the discovery of crystals with low nonproportionality.

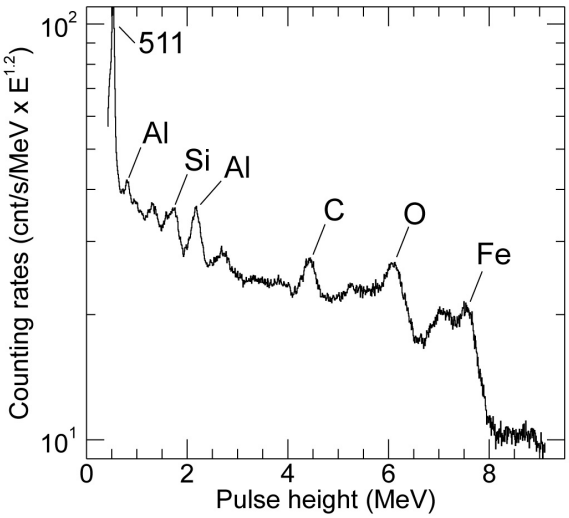

Figure 1. Gamma ray pulse height spectrum acquired by the NASA Dawn mission's Gamma Ray and Neutron Detector from low altitude at dwarf planet Ceres [5].

Detection efficiency is another key consideration in the design of planetary gamma ray spectrometers. Efficiency is determined by crystal size, geometry and gamma ray attenuation coefficient of the scintillating material. The latter increases with the density and effective atomic number (Z_{eff}) of the scintillator (Table 1). Bismuth germanate has the highest Z_{eff} of any scintillator used for planetary applications. Large, BGO crystals can be manufactured, resulting in very high detection efficiency.

New scintillators combine high light yields and low nonproportionality resulting in up to a factor of four improvement in resolution compared to BGO. Of these, $LaBr_3$:Ce is not well suited for planetary applications, despite having flight heritage [8]. Self-activity (radiolanthanum) obscures planetary gamma ray emissions up to 3 MeV. Strontium iodide, which yields

higher efficiency with equivalent energy resolution, and is therefore the focus of our research (Table 1) [9]. In addition, we are evaluating CHC for planetary applications. Its cubic structure will enable the growth of large crystals that are not hygroscopic. Substitution of Cs with Tl may yield scintillators with improved energy resolution and efficiency equivalent to BGO [10].

Testing and evaluation: Specific attributes of scintillators evaluated by our project include susceptibility to radiation damage, sources of background interference, and the response to gamma rays and energetic particles. In addition, we are optimizing the mechanical and optical design of scintillation detectors for space applications, including the evaluation of silicon photomultipliers for compact sensors deployed on microsatellites.

Preliminary radiation damage tests of SrI_2:Eu were carried out at the Loma Linda University Medical Center Proton Facility. Strontium iodide ingots were grown using the vertical Bridgman process by Fisk University. Small crystals ($10\times10\times10$ mm^3) were harvested from the ingots and polished, etched and encapsulated. Pre-irradiation tests included measurement of pulse height resolution and gamma-ray nonproportionality. The crystals were exposed to a range of doses, up to 20 krad in 100- and 200-MeV proton beams. Bismuth germanate crystals of the same size were simultaneously exposed. These served as reference that can be connected to doses received in flight. For example, BGO on Dawn was exposed to ~0.2 krad/yr during more than 10 years in deep space [11]. A noticeable loss in gain, likely due to darkening of the BGO crystal in response to radiation damage, was observed during flight. Nevertheless, performance of the sensor was acceptable through the end of the mission.

Post-irradiation characterization of SrI_2:Eu crystals revealed decreased gain with exposure, likely the result of damage induced darkening; although, neither differential nonlinearity nor energy resolution were significantly affected. Two-way transmission measurements using a spectrophotometer demonstrate darkening of the BGO crystals in visible to infrared wavelengths. The samples were placed in storage at room temperature for a long time prior to characterization. Thus, our preliminary experiments constrain the effects of deep defects introduced by exposure to protons.

Next Steps: Based on lessons learned, additional accelerator testing is planned to explore radiation damage effects and annealing of SrI_2:Eu and CHC crystals fabricated at Fisk University along with BGO crystals from commercial sources. The proton response of the scintillators will also be characterized. These studies will provide data needed for sensor design and to develop a models of damage effects useful for mission planning. The models will be validated against flight data from the Dawn mission. In addition, are developing prototype instrument for planetary applications using large volume (2" × 2" right circular cylinder) SrI_2:Eu scintillator, which will be ready for testing in 2019.

References: [1] Prettyman T. H., "Remote Sensing of Chemical Elements Using Nuclear Spectroscopy," *Encyclopedia of the Solar System (Third Edition)*, T. Spohn, T. Johnson and D. Breuer, eds., pp. 1161-1183: Elsevier, 2014. [2] Goldsten J. O. *et al.* (2007), *Space Science Reviews, 131*, 1, 339-391. [3] Cherepy N. J. *et al.* (2008), *Applied Physics Letters, 92*, 8, 083508. [4] Burger A. *et al.* (2015), *Applied Physics Letters, 107*, 14. [5] Prettyman T. H. *et al.* (2019), *LPS L*, Abstract #1356. [6] Cherepy N. J. *et al.* (2009), *IEEE Transactions on Nuclear Science, 56*, 3. [7] Valentine J. D. *et al.* (1998), *IEEE Transactions on Nuclear Science, 45*, 3, 512-517. [8] Zhu M. H. *et al.* (2013), *Sci Rep, 3*, 1611. [9] Prettyman T. H. *et al.* (2015), *SPIE Newsroom*, DOI: 10.1117/2.1201510.006162. [10] Fujimoto Y. *et al.* (2018), *Sensors and Materials, 30*, 7, 1577–1583. [11] Prettyman T. H. *et al.* (2003), *Nuclear Science, IEEE Transactions on, 50*, 4, 1190-1197. [12] Knoll G. F., *Radiation detection and measurement*: John Wiley & Sons, Inc., 1989.

Acknowledgements: This work is supported by the NASA Planetary Instrument Concepts for the Advancement of Solar System Observations (PICASSO) program, grant number NNX16AK42G.

Table 1. Selected properties of some inorganic scintillators. Data for light output and energy resolution are from [4, 6, 12]. Quoted values for resolution are for small crystals with optimized light collection and readout.

Scintillator	Density (g/cm^3)	Z_{eff}	MFP200* (cm)	Light output (10^3 photons/MeV)	Resolution (%FWHM 662 keV)
$LaBr_3$:Ce	5.1	45	0.76	60	2.6
SrI_2:Eu	4.6	50	0.70	110	2.8
Cs_2HfCl_6 (CHC)	3.8	54	0.68	54	3.3
Cs_2LiYCl_6:Ce (CLYC)	3.3	43	1.18	20	5
NaI:Tl	3.7	50	0.86	38	6.5
CsI:Tl	4.5	54	0.60	52	7
$Bi_4Ge_3O_{12}$ (BGO)	7.1	71	0.20	8.2	8

*MFP200 is the approximate mean free path of 200 keV gamma rays.