Miniature Neutron-Alpha Activation Spectrometer

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Abstract. We are developing a miniature neutron-alpha activation spectrometer for in-situ analysis of chem-bio samples, including rocks, fines, ices, and drill cores, suitable for a lander or Rover platform for Mars or outer-planet missions. In the neutron-activation mode, penetrating analysis will be performed of the whole sample using a γ spectrometer and in the α-activation mode, the sample surface will be analyzed using Rutherford-backscatter and x-ray spectrometers. Novel in our approach is the development of a switchable radioactive neutron source and a small high-resolution γ detector. The detectors and electronics will benefit from remote unattended operation capabilities resulting from our NEAR XGRS heritage and recent development of a Ge γ detector for MESSENGER. Much of the technology used in this instrument can be adapted to portable or unattended terrestrial applications for detection of explosives, chemical toxins, nuclear weapons, and contraband.

INTRODUCTION

We are developing a miniature neutron-alpha activation spectrometer (MiNAAS) for in-situ analysis of chem-bio samples, including rocks, fines, ices, and drill cores, suitable for a lander or Rover platform for Mars or outer-planet missions. In the neutron-activation mode, penetrating analysis will be performed of the whole sample using a γ spectrometer and in the α-activation mode, the sample surface will be analyzed using Rutherford-backscatter and x-ray spectrometers. The instrument is expected to provide composition over a wide range of elements, including the rock-forming elements, rare earths, radioactive elements, and light elements present in water and biological materials. The detectors and electronics will benefit from remote unattended operation capabilities resulting from our NEAR XGRS heritage and recent development of a Ge γ detector for MESSENGER. Much of the technology used in this instrument can be adapted to portable or unattended terrestrial applications for detection of explosives, chemical toxins, nuclear weapons, and contraband.

Novel in our approach is the development of a switchable radioactive neutron source (SRNS) and a small high-resolution γ detector (SHGD). The SRNS is based on the separation of α-emitting radioisotope material and light-element material that has a large α cross section for generating neutrons. When the α-emitter material is placed

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in close juxtaposition with the light element material, the neutron source is switched on. When the materials are separated, the neutron source is switched off, preventing unwanted activation and radiation damage to spacecraft and instrument components when measurements are not being made, without any shield. The SRNS does not have the bulky ancillary equipment required by an accelerator-based neutron source, such as an ultra-high voltage power supply (which tends to be relatively unreliable). The SRNS can provide a reasonably high neutron flux but is small, requires very little power, and is well-suited to remote unattended operation.

The SHGD consists of a small HPGe or CZT γ detector inside a CsI(Tl) or BGO anticoincidence cup, depending on resolution requirements and environmental conditions, along with custom signal processing electronics. The cup provides suppression of cosmic rays and detector γ scattering, along with an escape coincidence mode that further suppresses background at energies above 1 MeV, and also supplemental detection of high-energy γ lines.

INSTRUMENT DESIGN CONCEPT

The instrument must be suitable for a lander or Rover platform that meets mass, power, and environmental constraints of planetary missions. For Mars missions, a total mass of ~ 2-5 kg (including electronics) and an input power ~ 2-10 W is envisioned. For missions to the outer planets, a total mass of ~ 1 kg and input power of ~ 1 W would probably be necessary. There will be two operational modes, neutron activation for choosing samples and for bulk element analysis of each sample, and alpha activation for surface element analysis of each sample.

In neutron activation mode, the switching neutron source is on. If Be is used as the target material, neutrons up to 11 MeV will penetrate 10 cm or so of rock and react with nearly every element (primarily by inelastic scatter, some absorption), producing penetrating gamma-rays with energies characteristic of the element. Depending on interaction cross-section and element composition, one can detect rock-forming elements (Na, Mg, Si, Fe, Ca, etc), rare earths (Sm, Eu, for example), radioactive elements (K, Th, U), light elements in waters, ices, and bio-materials (C, N, O, and H by capture if there is neutron moderator present).

A conceptual design of the MiNAAS instrument is shown in Fig. 1, operating in the neutron activation mode. An alpha source is located in the center position opposite the target surface. An on/off neutron source is accomplished by moving a thin Be foil between the alpha source and the target. A small stepper motor drives the foil holder mechanism. This allows one source to provide alpha particles for the Rutherford backscatter and X-ray fluorescence spectrometers, and neutrons for the prompt gamma spectrometer. While in the alpha-only position, the gamma ray spectrometer can make needed measurements of the gamma-ray background. The gamma-ray detector does not need to be located near the source and therefore is conveniently placed on the side of the sensor head. This allows room for the BGO or CsI anticoincidence cup.

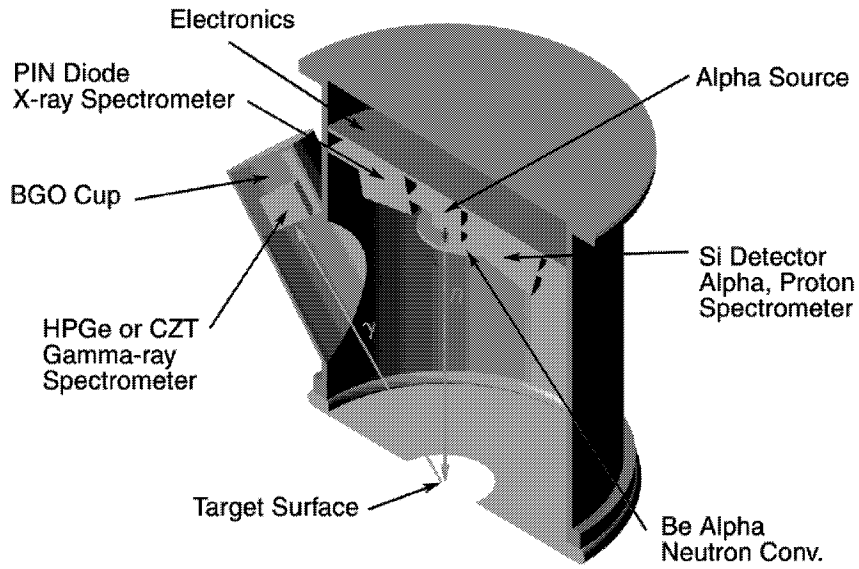

FIGURE 1. Conceptual design of MiNAAS instrument, shown in neutron activation mode.

Located close to the source is a small stack of surface-barrier solid-state detectors used for the backscatter and proton spectrometers, in the alpha activation mode. A thin front detector measures the energy of the backscattered alpha particles, allowing differentiation of light elements (C, N, and O). A thicker rear detector stops energetic protons that penetrate the front detector. In this way, the experiment can discriminate between alphas and protons. For protons, the coincident energies of the two detectors are combined to yield the total energy. The alpha/proton reaction can identify Na, Mg, Al, Si, and S.

For the X-ray spectrometer, a separate miniature Si PIN photodiode or drift detector measures characteristic x-rays in the 1-10 keV region. A thin Be window in front of the detector shields it from alphas and protons, yet is transparent to low-energy x-rays. X-rays in the sample excited by fluorescence from alphas and L-shell x-rays from the source allow identification of intermediate mass elements (Mg through Ni). Shown in Fig. 2 is the conceptual instrument design, operating in the alpha activation mode.

An initial penetrating scan of the planet surface by the gamma-ray spectrometer can examine soils, ices, loose materials, other objects, and major constituent elements, to choose samples from. Chosen samples can be analyzed in bulk and the source can then be switched to alpha activation mode for surface analysis of samples, eg. observing weathering layers, rinds, dust, or soil layers. The combination of the two complementary modes allows possible inferences into prebiotic conditions, petrology, planetary differentiation, igneous evolution, and weathering history.

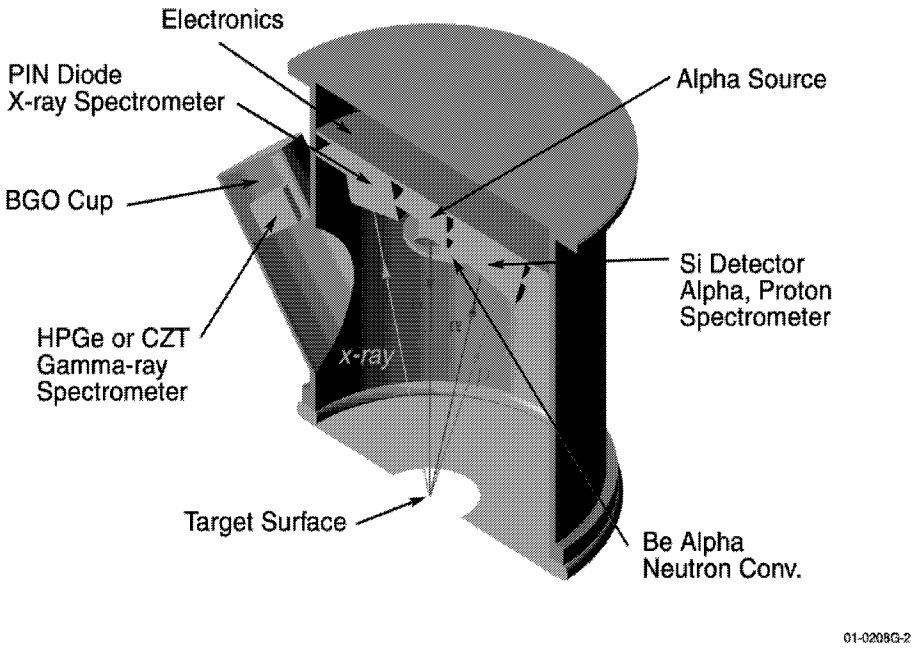

FIGURE 2. Conceptual design of MiNAAS instrument, shown in alpha activation mode.

SWITCHING RADIOACTIVE NEUTRON SOURCE

Exploration of planet composition and geology using neutron activation analysis requires a neutron source of adequate intensity in close proximity to the sample, that produces neutrons of sufficient energy to penetrate through the rock. Gamma rays characteristic of the elements will be provided by inelastic scattering, absorption, and moderation followed by capture of the neutrons inside the sample. However the source must be small in size and mass and require little power, in order to meet mission requirements for a Mars Rover or spacecraft exploring the outer planets. Also, the ability to switch the source on and off is highly attractive, to simplify pre-launch operations, avoid any activation of the spacecraft during flight, and allow sharing of an alpha source between neutron activation and APX spectrometries.

A switching neutron source satisfying these criteria can be developed, based on separation of alpha-emitting radioisotope material from target material consisting of specific isotopes of certain light elements, such as Be, B, and Li, that have high cross sections for alpha/neutron reactions. When these materials are separated so that the alpha particles do not strike the target, the source is switched off, and when these materials are brought together so that the alphas strike the target, the source is switched on. A possible geometrical concept for such a source is shown in Fig. 3.

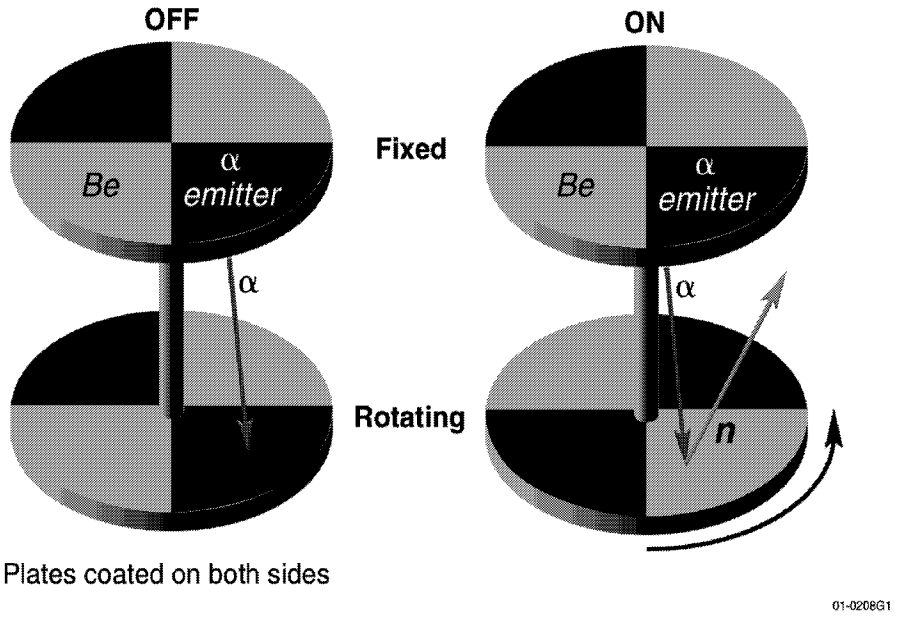

FIGURE 3. Possible geometrical concept for a switching radiosotope neutron source.

In an actual source, the plates would be located much closer together than shown in the figure, and raised ridges separating the segments would prevent alphas from crossing over to an adjacent plate. In Fig. 3, the alpha emitter is deposited to a depth equivalent to the alpha range; further depth would increase the background from reactions in the emitter material (such as gamma radiation, alpha/neutron reactions with impurities, and spontaneous fission) without a concurrent increase in the switchable neutron rate, lowering the on/off switching ratio. For a given alpha emitter and target, neutron intensity is increased by increasing the plate area or stacking pairs of plates.

Depending on the conversion rate of the alphas emitted in the layers to neutrons and the half-life of the alpha emitter, this Switching Radioisotope Neutron Source (SRNS) concept has the potential to provide a compact low-mass switching neutron source requiring only the minimal power to rotate the movable plates one segment, which can be done quite reliably and lends itself well to remote unattended operation. No massive shield is required, such as would be needed to "shut off" a conventional radioisotope source. The competing type of switching neutron source is an accelerator, which requires power for bulky ancillary equipment, as well as an ultra-high voltage supply (~ 100 kV for even a deuterium beam and tritium target), which has been shown to be unreliable in space applications.

The conversion rate of alpha particles to neutrons and the resultant neutron energy spectrum are basically determined by the choice of SRNS target material, Be, B, or Li as considered here and shown in Table 1 below. Be provides the highest neutron yield

and the highest neutron energy, up to 12 MeV, but there is a 4.43 MeV gamma from de-excitation of a ^{12}C state that occurs with ~ 80% probability. B provides an intermediate yield and an energy spectrum similar to a fission spectrum, while Li provides a low yield, minimal gamma radiation, and a low-energy spectrum more suitable for moderation to thermal energies.

TABLE 1. Properties of Selected SRNS Target Materials

Target	Be	B	Li
Avg. N Energy, MeV	4.5	2.7	0.5
Max. N Energy, MeV	12	5	1.5
Neutron Yield, %Be	100	25 – 50	0.8 – 5
Gamma Energy, MeV	4.43	2.31	negl.
Gammas per Neutron	0.8	0.07	negl.

The choice of alpha emitter basically determines the overall alpha rate and corresponding neutron intensity, as well as the period of source usefulness, through its alpha half-life, and the background when the source is in its off position, through its spontaneous fission half-life, alpha/neutron reactions with impurities, and gamma decay products. A selection of alpha emitters is shown in Table 2 below. In rows three through six, the alpha emitter is assumed to be deposited uniformly to a depth equal to the range of the highest energy alpha emitted. In row five, a simple transport code was used to determine the neutron yield per alpha, based on the experimental alpha/neutron cross sections of Be, B, and Li.[1] In row six, the "on/off" switching ratio of neutron intensity is based on the results of row five and the ratio of the alpha emitter alpha half-life to its spontaneous fission half-life times the average number of neutrons per fission.

TABLE 2. Properties of Selected SRNS Alpha/Neutron Sources

alpha emitter		Th-228	Cm-242	Po-210	Ac-227	Pu-238	Am-241
half-life		1.91 y	163 d	138 d	21.6 y	87.8 y	432 y
energy, keV		80-2600	44	800	50-870	44-766	60
gamma dose 1m, mr/hr-cm^2		17,000	0.73	0.45	230	0.003	0.63
neutrons per cm^2 × 10^4 "on"	Be	2100	1900	1200	190	5.9	1.1
	B	600	610	470	55	2.2	0.4
	Li	100	10	10	8.4	0.064	0.012
neutron "on/off" ratio	Be	high	41	high	high	1250	high
	B	high	14	high	high	460	high
	Li	high	2	high	high	15	high

In Table 2, "high" in row six means that the switching ratio is greater than 10^5 based on the calculation. Such a high ratio will not normally be attained due to alpha/neutron reactions and spontaneous fissions caused by the presence of impurity isotopes inside the alpha emitter layer. The neutron yield in column six for Pu-238 and a Be target has been verified by radiochemistry glove box experiments.[1]

Although Th-228 and Ac-227 can provide high neutron yields for usable half-lives and high switching ratios, Table 2 shows that the gamma radiation from their daughters makes these isotopes undesirable for most purposes. Cm-242 and Po-210 can provide high neutron yields with low gamma dose, but their half-lives are too short for most purposes, and the Cm-242 switching ratio is not very high. Although their neutron yields are only moderate, Pu-238 and Am-241 have long half-lives and can provide relatively high switching ratios with low gamma dose. Preliminary design considerations indicate that an SRNS of intensity greater than 10^6 n/s can be fabricated from Pu-238 or Am-241 and Be having a total mass that is a relatively small fraction of a kilogram, leaving plenty of mass allocation for a gamma-ray detection system.

MINIATURE HIGH-RESOLUTION GAMMA-RAY DETECTOR

An HPGe detector would provide outstanding energy resolution and efficiency, but would have to be cooled to ~ 90K to keep radiation damage from cosmic rays under control and avoid frequent annealing. The Applied Physics Lab experience in developing an HPGe spectrometer for the MESSENGER mission to Mercury, cooled by a minicryocooler and thermally isolated by a Kevlar string suspension and nested low-emissivity shields, indicates that a similar configuration may be feasible in the cold Mars environment, for a relatively large rover that would allow the added mass and power required. This configuration is probably not feasible for a small rover or a mission to the outer planets. A room temperature high-resolution semiconductor gamma-ray detector would be more amenable to these power and mass constraints.

Prompt neutron activation produces a multitude of characteristic gamma rays in the 0.1-10 MeV region, but the mean free path length for high-energy gammas in a moderately dense material is ~ 5 cm, a value inconsistent with the size of current room temperature high-resolution detectors. However, by focusing on the low-energy portion of the gamma ray spectrum (0.1-1.5 MeV), smaller, realizable detector volumes can be considered that achieve sufficient energy resolution to provide significant science return for most of the key elements (Na, K, Al, Ca, Mg, Mn, Si, Ti, Fe, Cl, Th, Nd, U, Sm, Eu, Gd).

Cadmium Zinc Telluride (CZT), a wide-bandgap semiconductor that requires no cooling, appears to be a good candidate detector material. A typical energy resolution of ~ 3% FWHM for ^{60}Co at 300 K is readily available. One of our co-investigators, Zhong He of University of Michigan, has obtained better than 2% FWHM for ^{137}Cs for a 1 cm^3 crystal [2] and has been getting similar results for crystals up to 1.5 cm x 1.5 cm x 1.0 cm [3], using a coplanar electrode to eliminate low-energy tailing and a correction method for electron trapping. Although the experimental results with the reference detector indicate that the presently attainable CZT energy resolution is adequate, the presence of closely spaced lines from other elements makes further

improvement in energy resolution strongly desirable. If radiation damage to CZT from cosmic rays turns out to be a serious problem, a future candidate room temperature high-resolution semiconductor detector could be HgI_2, which is much more robust against radiation damage.

Zhong He's unique depth-sensing technique can be used to form multiple coplanar-grid anodes on the anode surface of a single crystal to form larger detectors without sacrificing energy resolution. A 3 cm x 3 cm x 1 cm coplanar grid CZT detector may be feasible; it would be the world's largest CZT detector. Our proposed single detector could, in principle, be replaced by a three-dimensional array of CZT detectors in an effort to increase detector efficiency and to extend the resolution to a higher energy range, but this would require a substantial amount of processing electronics. Such an approach is considered too complex for a rover-based instrument and may be too massive, and is presently not being considered.

The signal-to-background ratio is dominated by the gamma ray continuum produced through Compton scattering in the detector, particularly for small detectors. An active cup surrounding the CZT would work in anti-coincidence to suppress the scattering, as well as help suppress cosmic ray background, help provide directionality to the CZT view field, and help protect the CZT from radiation damage and activation. Additionally, the high density and moderate energy resolution of the cup itself will provide sensitivity to some of the stronger high-energy gamma lines such as H (2.2 MeV), C (4.4 MeV), and O (6.1 MeV), that lie beyond the energy range of the CZT detector. A CsI(Tl) or BGO scintillator cup coupled to a PIN photodiode would eliminate a photomultiplier tube and its high voltage power supply.

RADIATION TRANSPORT COMPUTATIONS

We have used the MCNPX coupled neutron-gamma transport code to compute the gamma-ray flux per source neutron induced by an alpha-Be neutron source placed close to a rock of the "soil-free" composition of a Martian andesite, roughly 15 cm in radius, sitting atop Martian soil (composition from site A-2 of the Pathfinder mission [4]). The neutron source and gamma detector are both 15 cm above the soil, and ~1cm from the rock. The resulting gamma spectrum from the rock sample at the detector is shown in Fig. 4 for the low-energy region appropriate for a CZT detector. Multiple lines are seen from which to identify and quantify most of the major elements in the rock sample.

The gamma flux at the detector from the nearby Martian soil induced by the neutron source was found to be 50 to 70 times smaller than that from the sample, depending on the energy. But of further concern is the magnitude of the sample gamma flux compared to that induced naturally on Mars by cosmic ray bombardment. This latter gamma flux has been estimated using the Lahet and MCNP codes [5]. Assuming a neutron source of 10^6 n/s, the weakest neutron source we contemplate, the relative peak fluxes from the neutron source and from cosmic rays have been estimated and are shown in Table 3, which indicates that we will have signal peaks from 2 to 3 orders of magnitude larger than peaks produced by natural background.

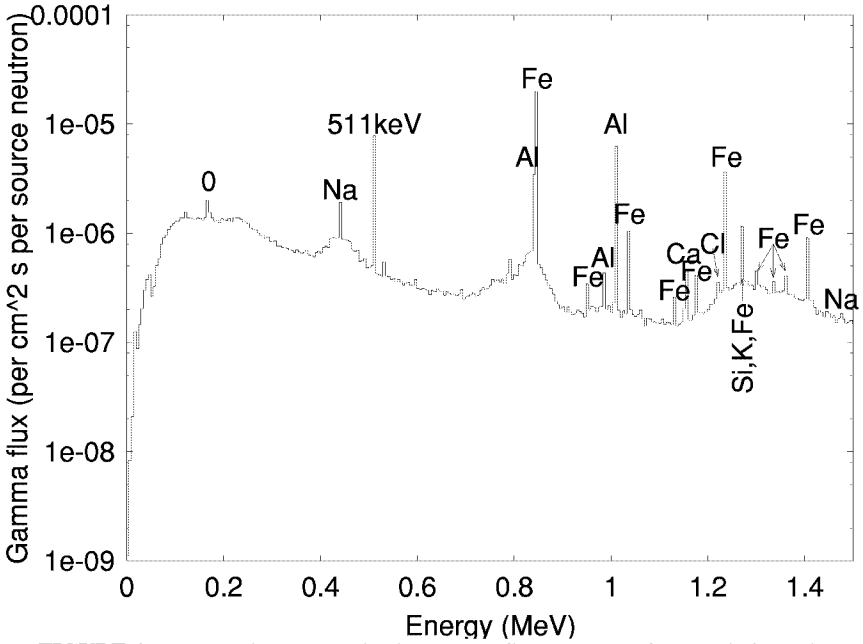

FIGURE 4. Computed neutron activation gamma flux spectrum of Mars adesite rock.

TABLE 3. Peak Gamma Counts in CZT Detector

target	w/o	Energy keV	GCR flux	α-Be flux	peak effic.	counts/s	hrs for 10^4 cnts
Na-23	1.93	439.9	0.0036	1.47	0.22	2.9	1.0
Al-27	5.61	1014	0.0055	6.11	0.06	3.3	0.8
Si-28	28.98	1779	0.0694	74.49	0.024	16.1	0.2
Ti-48	0.42	1037		0.949	0.058	0.5	5.6
Fe-56	8.57	846.7	0.0421	19.6	0.079	13.9	0.2

Also shown in Table 3 are the peak efficiency of a 3cm×3cm×1cm deep CZT detector, the resultant counts per second, and the number of hours to attain 1% Poisson count statistics, for key isotopes in the rock sample. An hour or less is required to accumulate 1% statistics for all isotopes shown except for Ti-48, which has a very low fractional composition. These computations do not consider background from the rover, gamma scattering inside the detector, or scattered gammas from cosmic rays. (In Table 3, flux is per cm^2/s).

The ultimate goal of our work is to determine composition from the uncollided prompt gammas, which will be detected as photopeaks and escape peaks in our gamma detector system. One of us, James Holloway, has recently studied the mathematics of the inverse problem of extracting composition information from large sample prompt gamma measurements [6] and has shown that the problem can be solved even in the presence of the significant complicating factors of neutron slowing down and thermalization in the unknown sample, and in the presence of nearby

materials. An iterative solution to the problem is based on repeated neutron-gamma transport computations in materials of estimated composition and known geometry, and results on the convergence of the iteration are in hand.

Recently [7] we have undertaken prompt gamma measurements of large samples of simple binary and ternary mixtures, irradiated with a 5Ci AmBe source. From these measurements, the composition of the mixtures was successfully reconstructed using the computation driven iteration, providing us with both experience and a proof-of-principle in large sample prompt gamma analysis measurements. Sample composition was successfully determined even in the presence of known interferences from other materials in the measurement setup.

REMOTE OPERATION, CALIBRATION, AND SAMPLING

The Applied Physics Laboratory has considerable experience with remote operation, calibration, and sampling of instrumentation through development and operation of many spacecraft and spacecraft instruments, including deep space missions and a number of nuclear spectrometers. The miniature neutron-alpha activation spectrometer will receive the benefits of this experience.

Critical parameters affecting operation and calibration of the instrument must be internally monitored, such as the temperature and voltage of power supplies, the temperature of preamplifiers, amplifiers, ADC's, and detectors, the leakage current of semiconductor detectors, and detector total counts. Based on values of the critical parameters, autonomous safing and recovery routines must be employed, as it is not feasible to perform these operations expeditiously from the ground, due to delays in communication with the spacecraft and slow telemetry rates over long distances.

Variable telemetry rates to accommodate mission objectives are quite useful, in conjunction with data accumulation internal to the instrument or spacecraft for indefinite periods. Disparities between sampling periods and strategies and telemetry delays and rates must be accommodated. Minimal data and housekeeping information need to be transmitted periodically, and autonomous event detection algorithms may be needed to record specific data (such as gamma-ray burst timing). Instrument software should be divided into "core" code and "application" code. "Core" code, that needs to be robust and unchangeable, includes diagnostics, error checking, communication, and boot routines. The remaining, "application" code should be uploadable and flexible, with new functionality easily added.

It is quite useful to have a complete terrestrial model that duplicates the flight instrument as closely as possible (usually the "engineering" model). This allows testing effects of new operations and software uploads, as well as post-launch calibration of many parameters of the flight instrument. Of course, the flight instrument must receive an adequate pre-launch calibration of parameters unique to it (temperature, INL, DNL, gain and discriminator settings, etc.). Sources available for energy calibration include natural radioactive sources in space (and on planets), on-board radioactive sources, and on-board electronic pulsers (though it is difficult to eliminate pulser temperature dependence). By choosing electronic, detector, and

mechanical components and operating conditions for long life with minimal aging, the instrument will remain in calibration for a relatively long time period.

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