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PGAA/NAA Analysis with the Lawrence Berkeley National Laboratory (LBNL) D+D Neutron Generator

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Abstract: *Prompt Gamma Activation Analysis (PGAA) at nuclear reactors has undergone a renaissance with the advent of modern spectroscopy equipment, development of a precise database for PGAA analysis, and the development of guided neutron beams and remote target facilities far from the reactor core. PGAA at these facilities is complemented by short-lived Neutron Activation Analysis (NAA) because decay gamma rays are observed either simultaneously with prompt gamma rays or separately if the neutron beam is chopped. Activities with half-lives as short as 1 ms can now be analyzed with NAA. Sensitivity of less than 0.1 mg/g of any element except Helium has been achieved with a 10^6 $ncm^{-2}s^{-1}$ thermal neutron beam. This kind of analysis has so far been limited to a handful of reactor facilities around the world. At Lawrence Berkeley National Laboratory (LBNL) we are developing a PGAA/NAA analysis system based on a compact, low power, »4´10^9 n/s D+D (E_n»2.5 MeV) neutron generator. The generator creates minimal gamma-ray background so detectors can be placed close to the target where the neutron flux is comparable to the guided neutron beam at a reactor. A D+D generator requires less thickness of moderator to thermalize neutrons than a conventional D+T (E_n»14 MeV) generator and eliminates the environmental and political concerns raised by using tritium. This paper discusses our initial PGAA/NAA experimental results for the LBNL neutron generator and future plans to develop reactor-quality PGAA/NAA analysis in the laboratory.*

I. INTRODUCTION

Neutron-induced Prompt Gamma-ray Activation Analysis (PGAA), also called Prompt Gamma Neutron Activation Analysis (PGNAA), is a nondestructive nuclear method for quantitative elemental analysis that was first demonstrated by Lea [1] and the Fermi group [2] in 1934. First measurements by reactor based PGAA were published in 1966 [3-5]. High-resolution germanium semiconductor detectors were first applied to neutron capture spectroscopy in 1967 [6], and to Neutron Activation Analysis (NAA) in 1968 [7]. The next major advance for PGAA analysis was the application of thermal and cold neutron beams from neutron guides [8] which made it possible to perform measurements in low background areas far from the reactor core. Lone *et al* [9] published the first comprehensive database of gamma rays for PGAA analysis in 1981. This database proved useful for qualitative analysis but had many mistaken assignments and erroneous cross sections making it less useful for quantitative analysis. A new PGAA database suitable for quantitative analysis to a precision of 1-3%

for all elements was measured at Budapest [10,11] and published by IAEA in 2003 [12]. PGAA can now be used to analyze <0.1 mg/g of any element except Helium [13]. An example of the analytical power of modern PGAA is shown in Table 1 where 25 elements were simultaneously analyzed in deep-sea vent samples collected by the ALVIN submersible [14].

Since quantitative PGAA is mainly restricted to a handful of nuclear reactors, it is desirable to find other neutron sources for this application. Although reactors have the clear advantage of high neutron flux for NAA, guided neutron beams of $<10^6$ $ncm^{-2}s^{-1}$ are used for PGAA. Spontaneous-fission sources like ^{252}Cf have too low a neutron flux and are too radioactive to be used for PGAA.   D+T ($E_n \approx 14$ MeV) neutron generators are compact and can produce 10^{10} ns^{-1} with no inherently radioactive "core". This is adequate to produce a useful neutron flux for PGAA analysis at a low background target position. D+T neutron generators have long been used for NAA [15] and more recently used for specialized applications of PGAA [16]. Particle accelerators have also been used to produce higher neutron fluxes for

PGAA [17], but these facilities tend to be large and expensive.

TABLE 1. Percent elemental concentrations (percent by mass) for the three ocean vent samples. Oxygen values marked with an asterisk were calculated from expected oxidation states for the observed elements.

	ALVIN 917-R4	ALVIN 1457-1R-C	ALVIN 1461-2R
O	45.9*	41(6), 44.9*	45.1*
S	20.0 (2)	0.151(5)	0.16 (1)
Ca	11.3 (2)	7.22 (11)	7.25 (13)
Fe	9.28 (11)	9.65 (8)	9.37 (9)
Cu	7.67 (7)	---	---
Al	---	7.10 (7)	7.06 (12)
Mg	1.8 (2)	3.98 (11)	3.6 (2)
Zn	1.36 (5)	---	---
P	---	0.85 (18)	1.6 (2)
Ni	1.17 (3)	0.022 (2)	---
Ti	---	1.097 (8)	1.060 (10)
Si	0.55 (5)	22.6 (3)	22.3 (3)
H	0.368 (4)	0.0290 (5)	0.027 (1)
K	0.27 (6)	0.138 (4)	0.16 (1)
Cl	0.194 (2)	0.0566 (5)	0.0188(5)
Mn	---	0.154 (2)	0.161 (4)
Na	0.140 (14)	1.97 (4)	1.96 (5)
V	---	0.042 (2)	0.046 (3)
Co	0.0066(11)	0.0045(3)	0.0058 (9)
Sc	---	0.0039 (2)	0.0058 (5)
Cd	0.00352(5)	---	0.00024 (3)
B	0.00220 (2)	0.000659 (7)	0.000658 (8)
Dy	---	0.00099 (8)	0.00111 (14)
Gd	0.000050 (6)	0.000524 (7)	0.000556 (10)
Sm	0.00033 (3)	0.000330 (5)	0.000340 (7)

Commercial D+T neutron generators have several drawbacks including loss of neutron flux with target depletion and safety and regulatory problems associated with tritium handling. D+D ($E_n \approx 2.5$ MeV) neutron generators would avoid the tritium issues, and provide neutrons that could be thermalized for PGAA measurements with less moderator. The D+D neutron production cross section is only $\approx 1\%$ of the D+T cross-section, so until now D+D neutron generators were seldom employed in applications. At LBNL we have developed a compact, high flux D+D neutron generator that has provided $>10^9$ n/s, comparable to D+T generators. The LBNL D+D generator has been described in a separate paper [18]. The generator has a self-loading target and operates indefinitely at constant neutron flux. In this paper we discuss the construction of a neutron generator facility at LBNL and report first experimental results of NAA and PGAA analysis with the LBNL D+D neutron generator.

II. EXPERIMENTAL RESULTS

Preliminary experiments have been performed at the LBNL Neutron Generator Facility to determine the efficacy of PGAA/NAA analysis with neutron generators.

II.A. LBNL Neutron Generator Facility

The LBNL neutron generator facility is shown in Figure 1. The neutron generator is surrounded by 30 cm of polyethylene that acts as both moderator and neutron
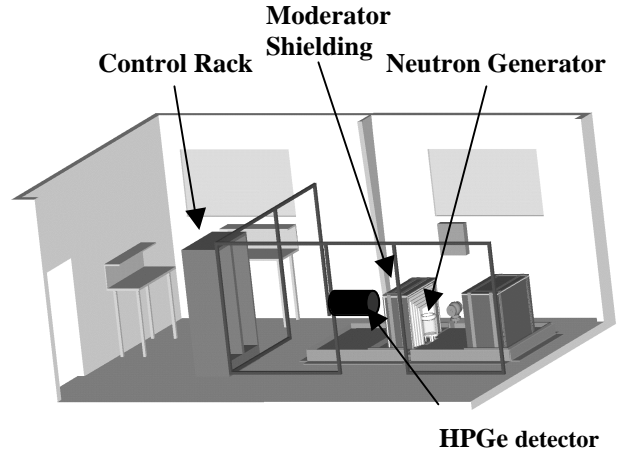

FIGURE 1. LBNL Neutron Generator Facility

shielding. Lead-lined plywood surrounds the moderator for gamma ray shielding. The moderator is divided so that it can be pulled apart for access to the neutron generator. A port with a removable plug is provided on the left side of the moderator in Figure 1 for the insertion of PGAA/NAA targets. A portable 20% efficient HPGe detector, shown facing the port in Figure 1, is used for both PGAA and NAA measurements. Data are acquired with an ORTEC Digital Signal Processor and analyzed with Hypermet PC software [19]. Energy and intensity calibrations to 2.75 MeV were performed using NIST standard sources and ^{24}Na, produced by the neutron generator. Calibrations up to 10 MeV were performed with prompt gamma rays as discussed below.

II.B. PGAA Experimental Results

Our initial PGAA spectrum from the LBNL Neutron Generator Facility, operating at $\approx 4 \times 10^9$ n/s, is shown in Figure 2. It was recorded with the detector directly facing the moderator plug. This is a PGAA background spectrum produced by the neutron generator and surrounding materials. The elements C and H (polyethylene), Ti (neutron generator target), Fe and Al (supporting materials), Si and O (concrete), Cl (plastics), and B (detector neutron shielding) are observed.
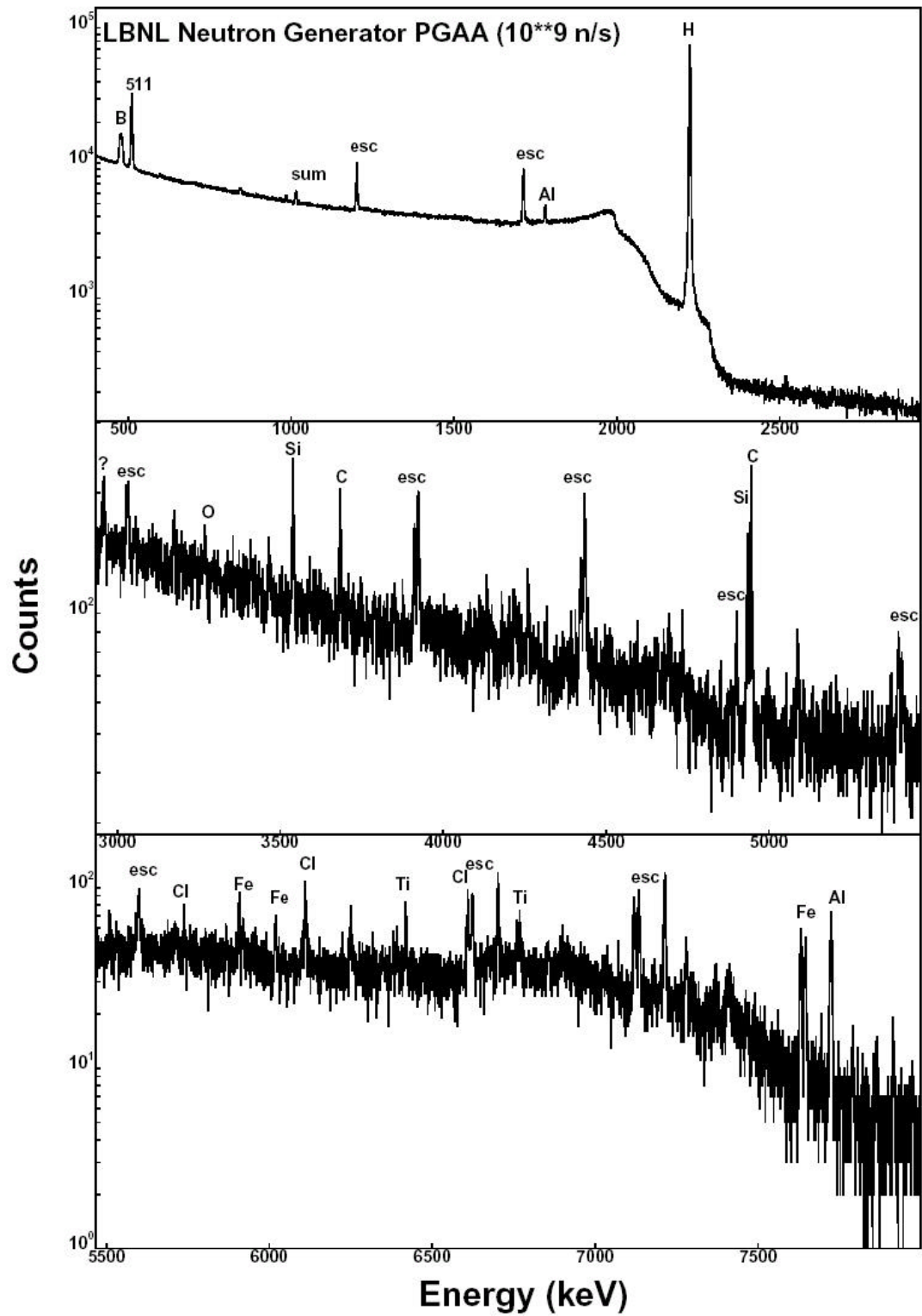
FIGURE 2. PGAA background spectrum counted for 15 minutes at 40,000 counts per second at the LBNL Neutron Generator Facility. The HPGe detector was positioned directly adjacent to the moderator plug , as shown in Figure 1, and wrapped in borated silicone rubber to avoid neutron activation.

This background count rate is very high and must be lowered before routine analysis can begin. Plans for reducing this background are discussed below.

II.C. NAA Experimental Results

Small targets for Neutron Activation Analysis at the LBNL Neutron Generator Facility are placed in a thermal bombardment chamber in the port plug and 3 cm of polyethylene is provided for neutron thermalization. We determined that the thermal neutron flux at the target position is $\approx 10^5$ ncm^{-2}s^{-1} by NAA with copper foils. Irradiated targets can normally be retrieved within 30 seconds after bombardment   Larger samples can be accommodated directly next to the neutron generator, but they require ≈ 1 minute for retrieval.

We are systematically measuring spectra for all applicable elements to determine our NAA detection sensitivity. Spectra have been obtained for Na, Al, Cl, Mn, Co, Cu, Ga, Br, Sb, I, and Hf. Figure 3 shows an NAA spectrum of cobalt where the 826.3- and 1332.5-keV gamma rays from ^{60}Co^m(t$_{1/2}$=10.5 min) are observed.
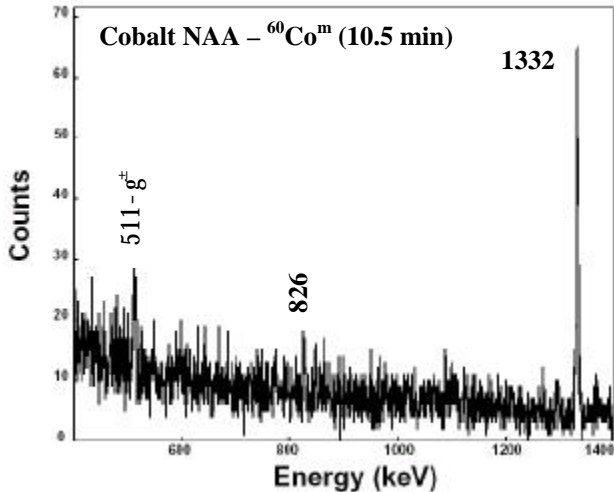

FIGURE 3. NAA analysis of 1 g sample of cobalt metal bombarded for 10 min and counted for 12 minutes.

Additional NAA experiments have been performed using 2.5 MeV neutrons without moderator. Figure 4 shows a gamma ray spectrum demonstrating analysis of Hg by the ^{199}Hg(n,n')^{199}Hg^m(t$_{1/2}$=42.6 min) reaction where the 158.3- and 374.1-keV gamma rays are clearly seen. Many isotopes that are not measurable by NAA with thermal neutrons can be analyzed by neutron-induced reactions. We have also developed a method to analyze uranium with 2.5 MeV neutrons. Figure 5 shows a comparison of gamma ray spectra for depleted uranium before and after bombardment. High-energy fission product gamma rays from ^{90}Rb^{m+g} (t$_{1/2}$=158 s + 258 s), ^{92}Rb(t$_{1/2}$=4.5 s), and other isotopes create a unique signal

that can be useful for uranium analysis and detection of concealed fissionable materials. Molnar *et al* [20] first demonstrated this method for thermal neutron induced fission of ^{235}U.
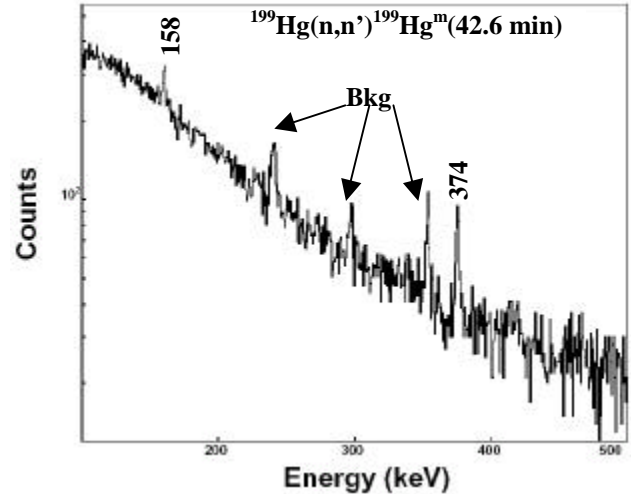

FIGURE 4. NAA analysis of 50 g sample of HgSO$_4$ bombarded for 30 min and counted for 30 min.
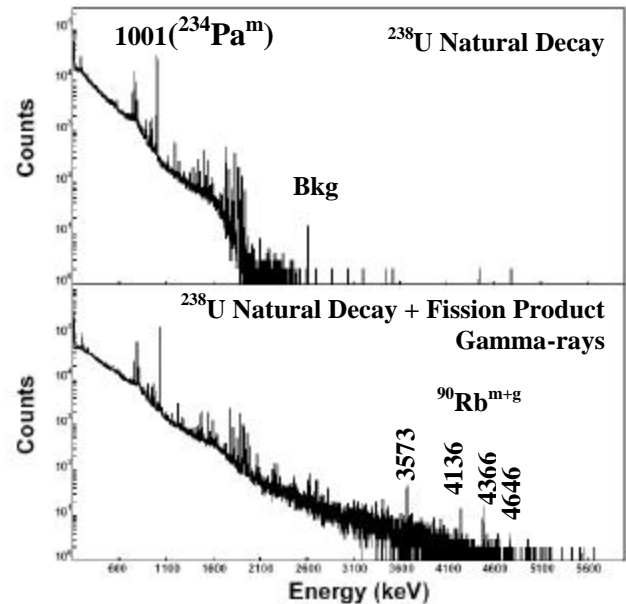

FIGURE 5. Detection of uranium by high-energy fission product gamma rays. The top spectrum records the decay of depleted uranium counted for one hour, and the bottom spectrum shows the same sample counted for 6 minutes following a 10-minute bombardment. The energies of gamma rays from the decays of ^{90}Rb^{m+g} are indicated.

III. FUTURE PLANS

These results demonstrate the feasibility of PGAA/NAA research with the LBNL D+D neutron generator. Considerable effort still remains to modify the facility in order to achieve more quantitative results.

III.A. PGAA Plans

Our goal is to develop a PGAA target position as close as possible to the neutron generator. Also, we want a detector position near the target without viewing either the neutron generator or moderator directly. The PGAA background spectrum shown in Figure 2 is dominated by the 2223-KeV gamma ray from hydrogen in the polyethylene moderator. It is difficult to shield the detector from high-energy gamma rays, so we propose using borated polyethylene that will significantly reduce the 2223-keV gamma ray intensity and produce mainly 470-keV gammas by $^{10}B(n,\alpha)$ that can be more readily shielded. Scattered neutrons near the detector can be absorbed with ^{6}Li-doped materials by the ^{6}Li$(n,\alpha)^3$H reaction that produces no gamma rays. Additional background reduction can be accomplished by lining the base of the generator with borated polyethylene to reduce prompt gamma rays from the concrete floor. By pulsing the neutron generator, and taking data only when neutrons are present, the contribution of room background to the spectrum can be reduced. We expect to achieve $>10^5$ ncm^{-2}s^{-1} at the target position which is $\approx$10% of that achieved with the old thermal guided neutron beam at the Budapest Reactor [21]. The gamma ray background is expected to be higher than that achieved at Budapest, and sensitivity to H, B, C, Al, and Fe, may be less.

At Budapest, a 25% Compton shielded detector was positioned 25 cm from the target. Compton suppression helps collimate the detector towards the target and greatly reduces background, particularly in the high-energy region. Recent investigations with HPGe clover detectors indicate a tenfold increase in detector efficiency, with respect to a single detector, for high-energy gamma rays, and 10% peak efficiency at 10 MeV [22]. Improved detector efficiency can compensate for lower neutron flux allowing us to achieve results comparable to those achieved in Budapest with their old thermal guided neutron beam.

III.B. NAA Plans

Although significant progress has already been made to demonstrate NAA with the LBNL neutron generator, sample-handling techniques can be improved. We plan to construct a fast pneumatic rabbit transport system to move samples rapidly from the bombardment area to a remote, low-background counting station. The NAA target bombardment chamber would have moderators configurable for thermal and high-energy bombardments. NAA of short-lived neutron capture products would occur at the PGAA target position by pulsing the neutron generator and detecting decay gamma rays between pulses. NAA sensitivity to half-lives of <1 ms can be attained. Standard coaxial HPGe detectors will be sufficient for most NAA experiments, and the LBNL Low-Background Counting Facility may be used for measuring long-lived activities.

III.A. LBNL Neutron Generator Development

The current D+D LBNL neutron generator is designed to produce 10^{10} n/s. Increasing the current and/or accelerating voltage, designing better target cooling, and providing more Ti target area can improve this. We envision increasing the D+D generator output ot 10^{12} n/s. The D+T reaction would increase this flux by another factor of 100 but requires a sealed generator design to avoid tritium release. The cold neutron flux at the Budapest Reactor is currently 5×10^7 ncm^{-2}s^{-1}, i.e. about 50 times larger their old thermal neutron flux [23], and a similar improvement can be achieved through LBNL neutron generator development.

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