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NGD cold-neutron prompt gamma-ray activation analysis spectrometer at NIST

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Abstract A new instrument for cold neutron prompt gamma-ray activation analysis is being designed as part of the recent expansion of the cold neutron guide hall at the NIST Center for Neutron Research. This instrument has a 10-fold increase in neutron flux and lower gamma-ray and neutron background compared to the PGAA instrument it will replace at NG7. Monte Carlo based simulation sofware and experimental setups are used in the design phase to mitigate background while preserving high neutron fluence. The new instrument will also provide more space for samples and for experimental setup compared to the old facility.

Keywords Prompt gamma-ray activation analysis · Neutron guides · Radiochemistry · Nuclear instruments

Introduction

Prompt gamma-ray activation analysis (PGAA) has been an important measurement technique at the NIST Center for Neutron Research (NCNR) since 1978. An instrument for thermal neutron (TN)PGAA, in operation since 1978 [1], was upgraded in 2000 to improve detection limits, increase sensitivity, and reduce the fast neutron and gamma-ray components of the beam [2]. This instrument has found important applications in the certification of H, B, and other elements in standard reference materials (SRMs). The addition of a cold source and neutron guides to the NCNR in 1990 created beams with significantly longer wavelengths ($\langle\lambda\rangle > 5$Å typically at most instrument locations in the newly-constructed cold neutron guide hall). A cold neutron (CN)PGAA instrument, located on the lower half of neutron guide NG7, came into operation in 199 [1, 3] and was upgraded in 1995 when the original D_2O ice cold source was replaced by a liquid hydrogen cold source [4, 5]. The CNPGAA instrument has better detection limits for hydrogen (<5 mg/kg in many matrices) and better sensitivity for several other elements than does the TNPGAA instrument. This is because the structural components and neutron shielding for this instrument are hydrogen-free and because of the larger capture cross-section for CNs. The instrument has therefore found application in the determination of low-level hydrogen and other elements in metals, semiconductor materials, and a wide variety of other materials [5, 6].

Several drawbacks are inherent in the NG7 prompt gamma instrument. A one-meter section of the upper half of NG7, which feeds the 30 m small angle neutron scattering (SANS) instrument, lies just 3 cm above the PGAA target irradiation position. The presence of this guide results in increased gamma-ray background, limits the size of samples that may be analyzed, limits space for installation of additional equipment, and prevents direct access to the target position while the reactor is operating. In response to these drawbacks, the PGAA is being relocated to a high-flux end-guide position. The current design work for the new instrument, specifics of the initial experimental configurations, and computational models are described in detail.

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Experimental

The new CN PGAA instrument is part of the expansion of the CN guide hall at the NCNR, which was completed in 2013. The expansion includes the addition of four new neutron guides, designated as NGA, NGBl and NGBu (upper and lower guides of NGB), NGC, and NGD, complementing the previously existing seven guides of the facility (Fig. 1). The new PGAA instrument is constructed at the end of neutron guide NGD, just downstream from a vertically-focusing guide section. Steel walls serve as shielding between PGAA and the adjacent NGC guide and as personnel shielding between the instrument and the workstation. The wall downstream from the instrument serves as a barrier between PGAA and the neutron depth profiling (NDP) instrument, which is scheduled to be moved there in 2016. The PGAA instrument will be operated initially in a simplified, temporary configuration described in this section. Proposed improvements for a more permanent configuration for the instrument are described later.

The PGAA instrument as initially constructed is far from optimal for neutron and gamma background. Features for significantly reducing both, including spurious signals due to leakage of TNs into the detector itself have been investigated using the Monte Carlo *N*-Particle (MCNP) code [7]. Selected configurations were then modeled and the calculations were experimentally verified for background measurements. The MCNP model for the temporary experimental setup of the instrument is as shown in Fig. 2. A spring-loaded, lead-lined [6] Li-glass local beam shutter located just upstream from the last section of neutron guide opens to emit a 2 cm × 2.7 cm beam of CNs through a 0.28 mm thick magnesium alloy window. A lead collar has been constructed around the guide exit to shield gamma rays emitted by the boron glass guide. The neutron beam travels through a temporary aluminum flight tube, lined inside with lithiated polymer and outside with borated polymer, and is collimated by a ≤2 cm circular aperture in [6] Li glass mounted just before the sample chamber. A second temporary lithiated polymer/borated polymer-lined aluminum flight tube is mounted just downstream from the sample, with a piece of enriched [6] LiF mounted at the end of this tube to serve as a beam stop. Samples are irradiated in an evacuable aluminum box (273 cm (h) × 171 cm (w) × 247 cm (l)) with removable magnesium alloy windows. The sample position is located 40 cm downstream from the guide exit. The box is lined with lithiated polymer and LiF tiles immediately in front of the detector position to prevent scattered neutrons from reaching the detector.

The sample chamber and flight tubes are mounted on an aluminum table, which also supports the detector assembly. A larger piece of [6] Li glass mounted on the downstream

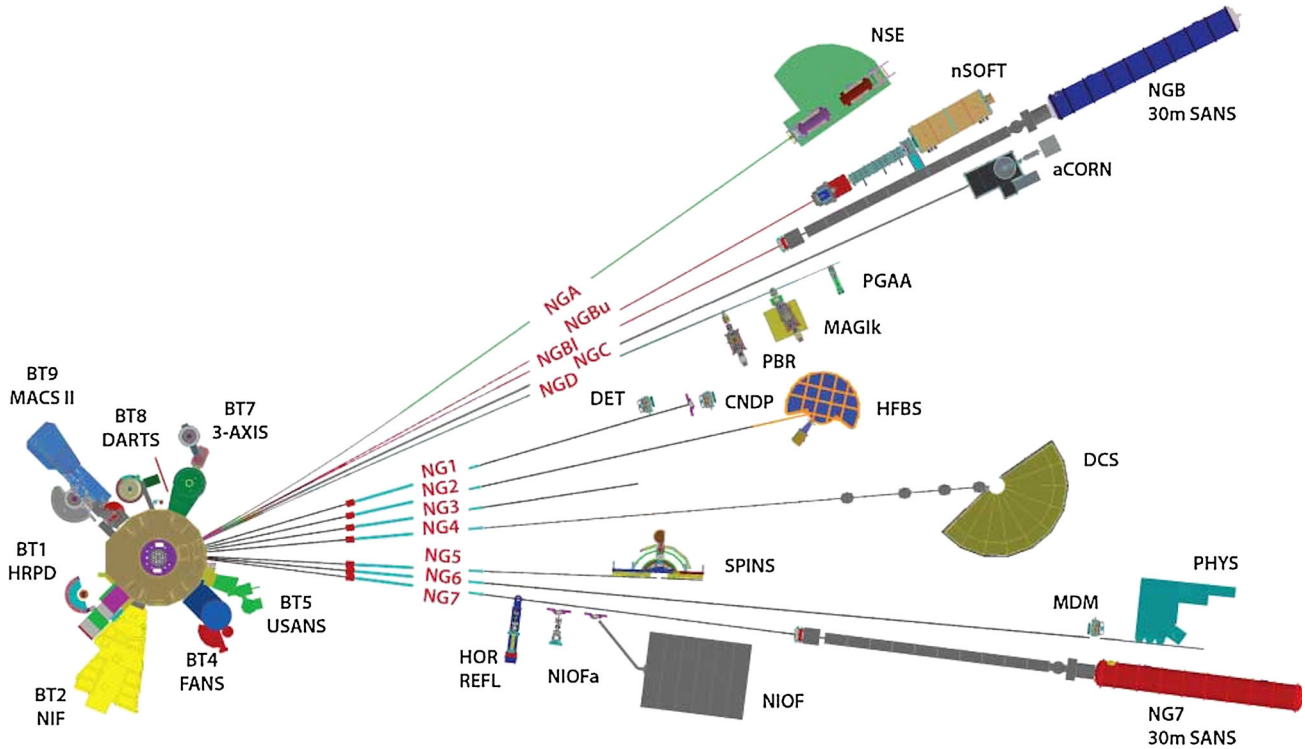

Fig. 1 Expanded cold neutron guide hall at the NCNR, showing the recently installed guides: NGA, NGBl and NGBu (*upper* and *lower* guides of NGB), NGC, and NGD

shielding wall, 79 cm from the sample box, serves as a permanent beam stop. Prompt gamma rays emitted by the sample are measured by a 41 % efficiency, 1.75 keV resolution, n-type Ge detector equipped with a transistor-reset preamplifier for high count rates. A bismuth germanate Compton shield will be installed later to improve signal-to-noise ratio. The detector is shielded on all sides by at least 10 cm (4 inches) of lead. Exchangeable lead collimators of diameters 1.27 cm (0.5 inch), 2.54 cm (1 inch), and 5.08 cm (2 inch) mounted in the front of the detector cave, allow control of the gamma-ray signal from the sample. The outside of the lead cave is covered by 0.318 cm (1/8 inch) thick cadmium to shield scattered neutrons, except in front of the lead aperture, which is shielded by a piece of lithiated polymer. The lead/cadmium shielded cave sits on a moving platform with its axis perpendicular to the neutron beam, with the detector end cap projecting through a hole in the bottom of the lead cave. The detector is mounted in a 30 L liquid nitrogen Dewar that sits below the detector table on a second moving platform. The upper moving plate is coupled to the top of the Dewar, allowing the entire detector assembly plus Dewar to be moved by means of a hand crank, thus enabling control of the sample-to-detector distance and additional control of the sample signal.

The detector is further shielded from gamma-ray background arising from neutron capture in the flight tubes and sample box walls by means of a wall of lead bricks placed between the sample box and the detector cave. This wall also shields capture gammas produced at the guide exit window and vacuum chamber windows. Gamma rays from the sample are admitted to the detector cave through a hole in the lead. Additional lithiated polymer inside the detector cave and borated polymer placed around the detector Dewar further serve to reduce gamma-ray background from capture of scattered neutrons.

Samples for analysis are normally sealed into bags of FEP Teflon and mounted in an aluminum frame suspended between Teflon threads. The frame is placed on a graph paper template to position the sample, and is then mounted inside the sample box. The optimum position of the sample in the beam was found by scanning a borax bead across the template to locate the position of maximum neutron flux.

A digital signal analyzer processes signals from the germanium detector. Gamma-ray spectra are collected on a computer using commercial gamma spectroscopy software.

Results and discussion

Measurement of neutron capture rate and background

Activation of a gold foil was used to measure the neutron fluence rate of the beam. The thermal equivalent neutron fluence rate was measured to be $9 \times 10^9\,\mathrm{cm}^{-2}\,\mathrm{s}^{-1}$ at the guide exit and $6.8 \times 10^9\,\mathrm{cm}^{-2}\mathrm{s}^{-1}$ at the NGD sample position, a 10-fold increase of the neutron fluence rate of $6.4 \times 10^8\,\mathrm{cm}^{-2}\,\mathrm{s}^{-1}$ measured in 2014 at the NG7 sample position.

Foils of titanium, iron, and copper, and a pressed disk of urea were used to measure element sensitivities at NGD. These measurements were performed using the temporary configuration shown in Fig. 2, at a sample-to-detector distance of 70 cm and using a 2.54 cm (1 inch) lead collimation of the gamma-ray signal. This configuration gives optimum gamma-ray signals for samples of a few hundred milligrams of most materials of interest. Table 1 gives sensitivities measured at NGD compared to sensitivities

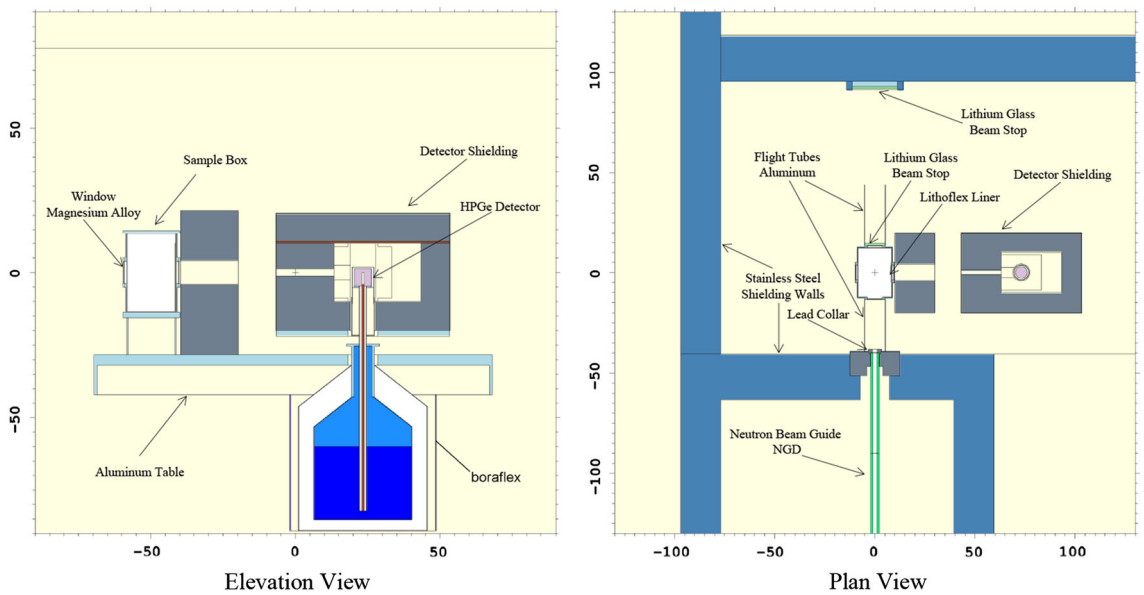

Fig. 2 MCNP model for the temporary setup of the new CPGAA instrument

measured at NG7, with a default sample-to-detector distance of 35 cm. A nearly two-fold improvement in sensitivity over NG7 was determined with the NGD instrument, despite the doubled sample-to-detector distance.

A background spectrum was collected for several hours by irradiating an empty Teflon bag inside the evacuated sample chamber. Count rates for elements measured in the background were converted to equivalent mass of element, measured at the sample position, using sensitivities measured for elements at NGD and estimated sensitivities for other elements. The results are given in Table 2 and are compared to background measured at NG7. The H background equivalent to 3.2 μg of H is half that measured at NG7. At NG7, the H background arises from three sources: neutron capture in atmospheric water vapor (essentially eliminated when irradiations are done under vacuum), capture by H in the graphite-based [6] Li paint applied as shielding to the adjacent upper NG7 guide (absent at NGD), and capture of scattered neutrons by H present in components in the germanium detector. The potential for significant additional reductions in background was identified from the MCNP simulations that showed that a major source originates from TNs scattered from the Mg alloy windows and from the sample. In the initial configuration, these neutrons were prevented from reaching the detector via a direct path by a piece of lithiated polymer, however, because the remainder of the sample box is unlined, neutrons scattered in all other directions traverse the aluminum walls of the sample box and are captured in the environment. The simulations clearly show that some of these neutrons are re-scattered from the liquid nitrogen dewar into the detector. This was evidenced experimentally by a germanium gamma ray signal and by an additional hydrogen peak from the carbon fiber composite detector window, which were reduced by factors 3 and 1.3, respectively, when a sheet of borated rubber was loosely draped around the liquid nitrogen dewar.

Table 1 Sensitivities (counts/s mg) measured for selected elements at NGD and NG7

Element/Energy (keV)	Sensitivity (cps/mg)	
	NGD (70 cm)	NG7 (35 cm)
Ti (1,381)	9.51	5.42
H (2,223)	20.1	12.1
Fe (7,631/7,645)	0.358	0.194
Cu (7,914)	0.192	0.117

Table 2 Background comparison: NGD versus NG7

Element (Energy keV)	NGD count rate element (cps)	Equivalent μg element (NGD)	Equivalent μg element (NG7)
H (2,223)	0.064	3.2	7.9
Ge (596)	0.75	400	800
Al (1,777)	0.17	280	490
Si (3,539)	0	0	3,900
Pb (7,370)	0.037	3,700	3,000
Cu (7,914)	0	0	140
Cd (558)	0.54	0.3	0.2
Fe (7,645)	0.015	42	380
B (478)	0.67	0.055	0.04

Future optimization of instrument and background

The addition of neutron flight tubes and judicious placement of neutron shielding plays a large role in optimization of background for this instrument (Fig. 3). The MCNP calculations have been used in the optimization of shielding and instrument setup. Several improved models have since been designed using MCNP modelling and are being analyzed. According to the calculations, the following improvements were accepted for future modifications. The interior of the sample box will be covered with a minimum of 5 mm thick lithiated glass (K2959) in 4π except at the

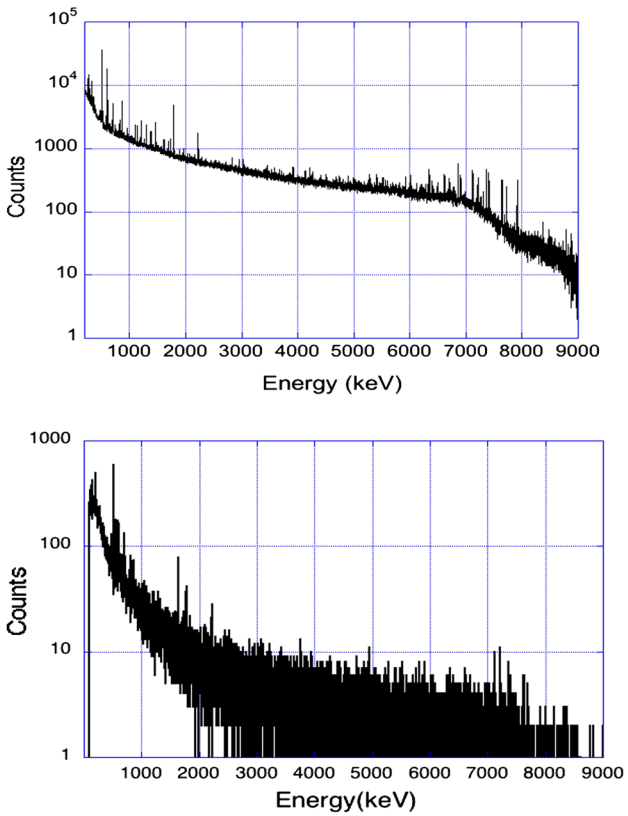

Fig. 3 Gamma-ray background spectrum measured at NGD, before (*top*) and after (*bottom*) installation of temporary neutron flight tubes and improved neutron shielding

beam entrance and exit. The detector penetration into the Pb cave will be additionally shielded with lithiated polymer and a lead plug. The temporary neutron flight tubes will be replaced by evacuable flight tubes, mounted directly on the sample box after removal of the sample chamber windows, allowing the box and flight tubes to be evacuated as a single unit and eliminating scattering and H capture in the air paths. To further reduce neutron scattering, the windowless tube downstream from the sample box will be extended to or beyond the end shield wall, with the beam stop relocated to the far end. Lead will be placed around the guide exit and vacuum chamber entrance to attenuate capture gammas from the windows.

Conclusions

The new instrument yields greater applicability, higher neutron flux, and better signal-to-noise ratio than the NG7 instrument. Future improvements will include optimization of neutron and gamma-ray shielding and installation of permanent evacuable neutron flight tubes. The improvement in sample space will allow the mounting of larger samples, and will also allow installation of equipment for specialized measurements. These include, but are not limited to, automated scanning stages for compositional mapping of samples, an automatic sample changer, atmosphere/temperature controlled sample chambers for studying in situ reactions, and additional detectors for performing coincidence measurements.

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