Cold neutron PGAA facility developments at university research reactors in the USA

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The PGAA applications can be enhanced by using subthermal neutrons, cold neutrons at university research reactors. Only two cold neutron beam facilities were developed at the U.S. university research reactors, namely at Cornell University and the University of Texas at Austin. Both facilities used mesitylene moderator. The mesitylene moderator in the Cornell Cold Neutron Beam Facility (CNBF) was cooled by a helium cryorefrigerator via copper cold fingers to maintain the moderator below 30 K at full power reactor operation. Texas Cold Neutron Source (TCNS) also uses mesitylene moderator that is cooled by a cryorefrigerator via a neon thermosiphon. The operation of the TCNS is based on a helium cryorefrigerator, which liquefies neon gas in a 3-m long thermosiphon. The thermosiphon cools and maintains mesitylene moderator at about 30 K in a chamber. Neutrons streaming through the mesitylene chamber are moderated and thus reduce their energy to produce a cold neutron distribution.

Introduction

The research applications at university research reactors can be enhanced by using subthermal neutrons, cold neutrons. The temperature of a neutron beam can be lowered by passing it through a moderator that is cooled. Neutrons with sufficiently long wavelengths (cold neutrons) can be reflected from some surfaces and they can be guided down cylinders, wave guides, without the normal $1/r^2$ attenuation and can be bent out of the lineof-sight paths followed by other radiation. Only two cold neutron beam facilities were developed at the U.S. university research reactors, namely at Cornell University and the University of Texas at Austin. The Cornell Cold Neutron Beam Facility (CNBF) included a moderator, a cryorefrigerator, copper cold fingers, a neutron guide system, vacuum jackets, shielding, and various connecting and control lines. The mesitylene moderator in the CNBF was cooled by a helium cryorefrigerator via copper cold fingers to maintain the moderator below 30 K at full power reactor operation. Cold neutrons from the mesitylene moderator were transported to an experimental facility using thirteen 1 m long natural Ni coated neutron guide elements. Texas Cold Neutron Source (TCNS) uses mesitylene moderator that is cooled by a cryorefrigerator via a neon thermosiphon. The operation of the TCNS is based on a helium cryorefrigerator, which liquefies neon gas in a 3m long thermosiphon. The thermosiphon cools and maintains mesitylene moderator at about 30 K in a chamber. Neutrons streaming through the mesitylene chamber are moderated and thus reduce their energy to produce a cold neutron distribution. The cold neutrons are transported out of the biological shield of the reactor and to a sample chamber location by a 6-m long curved neutron guide and an 80-cm long converging neutron

guide. Neutronic performance of both systems is known and published before. However, thermal and thermalhydraulic behavior of the both systems had never been analyzed. The investigation of thermal and thermalhydraulic characteristics of the cooling systems of both CNBF and TCNS are being carried out in order to design and build a third generation mesitlylene based cold neutron source at the Penn State University. After the characterization of the thermal behavior of both systems via analytical modeling and Computational Fluid Dynamics (CFD) solutions with $FLUENT¹$ and ANSYS2 codes, a model will be developed for the best performance of a university reactor based cold neutron source. The results of this investigation will be published later. The design features, cooling and warmup characteristics, and the performance of both CNBF and TCNS will be discussed here. Some measured PGAA results of TCNS system will be presented.

Background

Beams of neutrons produced by nuclear research reactors are used in condensed matter research to study the arrangement and interactions of atoms in materials. Because neutrons are highly penetrating, it is possible to probe deep within materials to characterize micropores, microcracks, small precipitates, polymers, biological macromolecules, etc. Neutrons examine matter at the atomic-scale in a way unmatched by other techniques because of their favorable wavelength to energy ratio. In many applications, the structural information provided by neutrons cannot be obtained in any other way.

A neutron beam can also be used for analytical analysis by neutron-capture gamma-ray spectroscopy. Concentrations of various elements in a sample can be determined from the measured emission rates of characteristic prompt gamma-rays produced by neutron capture. While neutron activation analysis is more sensitive for the determination of most elements, it cannot be used for some elements that can be analyzed by neutron-capture gamma-ray spectroscopy. In addition, basic nuclear physics studies are possible through neutron-capture reaction measurements of gamma-rays and internal conversion electrons.

All of the above research applications can be enhanced in one way or another by using subthermal neutrons, cold neutrons. Cold neutrons can be defined as neutrons with energies below 5 meV and corresponding velocity and wavelength 980 m/s and 4 Å, respectively. Cold neutrons have longer wavelengths and lower kinetic energies on the average than thermal neutrons, the majority of the neutrons normally present in neutron beams from nuclear research reactors. The temperature of a neutron beam can be lowered by passing it through a moderator which is cooled. To be effective, however, the temperature of the moderator must be significantly below liquid nitrogen temperature, 77 K.

Neutrons with sufficiently long wavelengths can be reflected from some surfaces in the same way light can be reflected from the interface between two transparent media. Cold neutrons, then, can be guided down cylinders, wave guides, without the normal $1/r^2$ attenuation and can be bent out of the line-of-sight paths followed by other radiation. Using long wavelength neutrons allows increased size scale for material structure research.

The usual neutron beams from a research reactor are contaminated by fast neutrons and gamma-rays that originate in the core. Filters, collimators, and shielding can be used to reduce these undesirable components to some extent. However, cold neutron beams can have a much lower gamma and fast-neutron background. Thus, detectors for capture-neutron and basic physics experiments can be placed closer to the sample, increasing sensitivity and making coincidence techniques feasible in many more situations.

Cornell Cold Neutron Beam Facility

Cornell Cold Neutron Beam Facility (CNBF) was located at one of the radial beam port of the 500 kW Triga research reactor and adjacent beam floor area (Fig. $1a)^{3-6}$ (Cornell University administration decided to close the Ward Center for Nuclear Sciences on June 2002, hence, Cornell reactor and Ward Center for Nuclear Sciences are no longer available for scientific community). The CNBF consisted of a cooled moderator, a cryorefrigerator, a copper rod (cold finger), and neutron guide elements (Fig. 1b). The moderator placed in a neutron beam port close to the reactor core. The moderator used in the Cornell source is mesitylene, a 1,3,5-trimethyl benzene. Because mesitylene freezes at 228 K and boils at 437 K, it is safer and much simpler to use than liquid hydrogen, D_2O ice, or solid methane, the more traditional cold-neutron-source moderators. The handling system for mesitylene does not need to withstand large or abrupt changes in pressure, but must be a closed system to avoid contaminating the mesitylene or releasing it since it is slightly carcinogenic and toxic.

The moderator was contained in a thin-walled aluminum right-circular cylinder 7.5 cm diameter by 2.5 cm deep position inside a beam tube at the graphite reflector of the reactor. The moderator was cooled by conduction through a $5-9$'s purity (99.999%) 1.8 cm diameter, 216 cm long copper rod. The copper rod was connected to the second stage of a cryogenic refrigerator located outside the biological shield of the reactor. A Gifford-McMahon cycle Cryomech Model GB04 helium cryorefrigerator was used for cooling. A typical measured cooling down and warming up temperatures are shown in Fig. 2. The moderator chamber temperature varied from 11 K at 0.0 kW reactor power with an evacuated chamber to a 28.5 K at 500 kW reactor power with a mesitylene filled chamber (Fig. 3). The neutron guide of Cornell CNBF contained thirteen 1-meter long elements. Each element was comprized of two and bottom by epoxied, ground glass strips of dimensions 2 cm wide by 100 cm long by 1 cm thickness. The cross sectional view resembled a "double" bar H", with internal dimensions of 2 cm wide by 5 cm high. The four interior surfaces were coated with a 5 Å thick evaporated layer of natural nickel. The predicted thermal equivalent flux at the exit of the neutron guides at 480 kW reactor power was about 4.10^6 n·cm^{-2.}s⁻¹.

Texas Cold Neutron Source

The operation of the TCNS is based on a helium cryorefrigerator, which liquifies neon gas in a 3-m long thermosiphon.^{7–12} The thermosiphon cools and maintains a cold neutron moderating material (mesitylene) at about 30 K in an aluminum chamber located inside the graphite reflector of the UT 1000-kW research reactor. The cooling down and warming up trends of the TCNS is similar to Cornell Cold Neutron Source. A typical measured cooling down temperatures of TCNS is shown in Fig. 4. Neutrons streaming through the mesitylene chamber are moderated and thus reduce their energy to produce a cold neutron distribution. The cold neutrons coming from the mesitylene chamber are transported out of the biological shield of the reactor and to the PGAA sample chamber location by a 6-m long curved neutron guide and an 80-cm long converging neutron guide. Figures 5a–c are a cross sectional view of the external components of the TCNS, curved and converging guides and the UT-PGAA facility. The

curved neutron guide is made up by three 2-m long sections, curved to a 300-m radius and divided into three vertical channels $(5\times0.45 \text{ cm}^2)$ by 0.1-cm thick walls. This array provides blocking of the straight-path background components streaming through the guide. The TCNS curved neutron guide, with all reflecting surfaces coated by a 1000- \AA ⁵⁸Ni layer, utilizes total reflection to transport neutrons without the normal $1/r^2$ intensity loss. The critical angle for total reflection of neutrons from 58Ni is 0.12° per Å. The characteristic wavelength of the curved neutron guide is 2.7 Å, which corresponds to neutron energy of 11 meV.

Taking advantage of the enhanced wave properties of cold neutrons, a converging neutron guide was added to the end of the final section of the curved neutron guide. The converging neutron guide was designed by JONG-YOUL KIM, K. ÜNLÜ, B. W. WEHRING and after examining several methods for neutron focusing and consists of four truncated rectangular cone elements. 13 Each element consists of four 20-cm long single-crystal Si plates coated with NiC-Ti supermirrors. The effective

critical angle of the converging neutron guide is $0.3^{\circ}/\text{\AA}$, providing a neutron reflectivity greater than 95%.

The combined effect of the TCNS cold neutron guide and focusing system can be summarized by the measured values of the neutron flux at the PGAA sample position. These measurements were done at low reactor power and the results extrapolated to 1000 kW. The resulting flux values are $(9\pm 2) \cdot 10^6$ n·cm⁻²·s⁻¹ with no moderator in the chamber (empty chamber operation) and $(1.5\pm0.2)\cdot10^7$ n·cm⁻²·s⁻¹ with the moderator (mesitylene) cooled to 26 ± 2 K in the chamber (cold moderator operation). For the case of empty chamber operation, the curved neutron guide selects neutrons on the low-speed side of the room temperature neutron distribution (calculated average speed of 1500 m/s and thermal equivalent flux of $(1.5\pm0.6) \cdot 10^7$ n·cm⁻²·s⁻¹). For the cold moderator operation, neutrons are selected from the cold-neutron distribution (calculated average speed of 800 m/s and thermal equivalent flux of $(4.6\pm0.7)\cdot10^7$ n·cm⁻²·s⁻¹).

Fig. 1. Cornell Cold Neutron Beam Facility layout (a), Schematic drawing of Cornell Cold Neutron Source (b)

Fig. 2. Typical cooling down and warming up (temperature vs. time) of Cornell Cold Neutron Source from room temperature to equilibrium operating conditions and operating temperature to room temperature

Fig. 3. Performance of Cornell Cold Neutron Source showing temperature versus varying reactor power. Mesitylene chamber temperature was about 12 K at 0.0 kW and increased to 29 K at 480 kW reactor power due to gamma heating of the chamber

Fig. 4. Cold head of cryorefrigerator and mesitylene moderator chamber cooling down behavior of Texas Cold Neutron Source

Initial PGAA Setup at UT

The UT-PGAA facility consisted of a sample chamber, a gamma-ray spectrometer with data acquisition and processing electronics, detector shielding, and a beam stop. $8-11$ During operation, the TCNS cold neutron beam illuminated the sample inside the PGAA chamber. This chamber consisted of an aluminum frame with a 14.5×14.5 cm² base and 16 cm of height, which sustained a 5-mil thick fluoro-ethylenepropylene (FEP) bag. The FEP bag was held to the base plate with an o-ring pressed by an adjustable aluminum frame. The base plate was provided with two tubes for purging the sample chamber with helium during operation and eliminate the background due to neutron captures in air.

The UT-PGAA gamma-ray spectrometer was based in a 25% efficient high purity germanium detector (GMX-25190-s Ortec) in the DUET configuration with an offset port Dewar (30 liters). This detector was selected in order to incorporate a Compton suppression system at a later date. The detector shielding was provided by a layer of 6LiF/poly (PNPI USA Corp., Houston, Texas) around the detector's crystal and a close pack lead cave. The close pack lead cave, built around the detector, has approximately 10 cm thickness except for the bottom side. The lead thickness at the bottom side was about 25 cm. A gap of about 2 cm was left between the 6LiF/poly detector wrap and the inner side of the lead cave. The detector sees the sample through a 6.5-cm diameter opening in the lead cave, shielded with a 6LiF/poly disk. For the initial arrangement, the sample–detector distance was 26.5 cm. This distance can be reduced with a different lead shield configuration. Layers of 6LiF/poly shield the front and the side faces of the lead cave and also the lateral sides of the sample chamber. A gadolinium converter plate in a neutron imaging system was used as a beam stop.

Performance of UT-PGAA

The initial step for testing and optimizing the UT-PGAA Facility consisted in using the empty chamber TCNS operating mode, the close pack lead cave detector shielding and 25.6 cm for the sample–detector distance. Samples of semiconductor wafers were positioned at 45° to the neutron beam axis. The size of the beam at the sample point is $\sim 1 \times 2.5$ cm². The amount of sample materials are determined by using the beam area. Since neutron flux is not uniform across the area of the beam and peaks at the center of the sample, a "small sample" area was also used for the estimated values of sensitivities (called small sample sensitivities).

For the initial performance test a borophosphosilicate glass on a silicon wafer and a thin nitride film deposited on silicon wafer were used. The borophosphosilicate glass sample on a silicon wafer was used for boron and silicon measurements, while the thin film nitride deposited on silicon wafer was used for hydrogen measurements. A segment of the measured gamma-ray spectra for a borophosphosilicate glass on a silicon wafer is shown in Fig. 6. The initial results for analytical sensitivities of boron, silicon and hydrogen target materials are presented in Table 1. The large sample sensitivities and detection limits for boron and silicon were based on our measurements in empty chamber operation mode (no cold moderator in the TCNS system and 1 MW reactor power). The sensitivity number reported for hydrogen was scaled from YONEZAWA's measurements at JAERI,¹⁴ while the detection limits were calculated using our measured background data.

For each element B, Si, and H the sensitivities and detection limits are reported for cold moderator operation mode and an optimized system using our estimated values. These estimations are based on measured neutron intensity values for empty chamber and cold moderator operation modes and assuming a 14.5-cm sample-detector distance.

Fig. 5. Cross-sectional view of the Texas Cold Neutron Source in the piercing beam port of the UT-Triga research reactor, showing the location of the 6 m long curved neutron guide and the 80 cm long converging neutron guide. All reflecting surfaces of the curved guide are coated with ⁵⁸Ni. Some details of converging neutron guide is shown in the right corner. The NiC-Ti supermirror layers coats all reflecting surfaces of the 16 Si single-crystal plates. A cross section of curved neutron guide is shown for comparison

Discussion

The UT-PGAA Facility sensitivities for boron, silicon, and hydrogen were measured to be 2300 cps/mg, 21 cps/g, and 5 cps/mg, respectively. Corresponding detection limits were 15 ng for boron, 0.5 mg for silicon,

and 2.5 µg for hydrogen. As to date, the sensitivities for other elements in various samples were not determined at UT-PGAA System. Only preliminary measurements were carried out at the end of 5 m section of 13 m neutron guides at Cornell University.

Fig. 6. A segment of the gamma-ray spectrum showing the ¹⁰B peak for a 3.8 µg sample. The results were obtained by the present, nonoptimized PGAA system with no cold moderator in the TCNS. In this situation, the curved neutron guide selects subthermal neutrons with an average speed of about 1500 m/s

Boron target material	Sensitivity, cps/mg	Detection limit, ^a ng
Empty chamber – present system		
Large sample $(>1.0 \times 2.5 \text{ cm}^2)$	290	120
Small sample $(0.5 \times 0.5 \text{ cm}^2)$	720 ^b	48 ^b
Cold moderator - optimized system		
Large sample $(>1.0 \times 2.5 \text{ cm}^2)$	900 ^b	38 ^b
Small sample $(0.5 \times 0.5 \text{ cm}^2)$	2300 ^b	15 ^b
Silicon target material	Sensitivity, cps/g	Detection limit, ^a mg
Empty chamber – present system		
Large sample $(>1.0\times2.5 \text{ cm}^2)$	2.6	4.2
Small sample $(0.5 \times 0.5 \text{ cm}^2)$	$6.5^{\rm b}$	1.7^{b}
Cold moderator - optimized system		
Large sample $(>1.0 \times 2.5 \text{ cm}^2)$	8.4 ^b	1.3^{b}
Small sample $(0.5 \times 0.5 \text{ cm}^2)$	21 ^b	$0.5^{\rm b}$
Hydrogen target material	Sensitivity, cps/mg	Detection limit, a μ g
Empty chamber – present system		
Large sample $(>1.0 \times 2.5$ cm ²)	0.6 ^b	20
Small sample $(0.5 \times 0.5 \text{ cm}^2)$	1.5^{b}	8 ^b
Cold moderator - optimized system		
Large sample $(>1.0\times2.5 \text{ cm}^2)$	2 ^b	6 ^b
Small sample $(0.5 \times 0.5 \text{ cm}^2)$	5 ^b	2.5^{b}

Table 1. UT-PGAA Facility performance

 a^{s} m = 3.29 $\sqrt{\frac{Bkg}{time}}$ /sensitivity, time = 4hrs.

b Estimated values.

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Table 2. Reactor based PGAA facilities

Table 3. List of references for each facility provided

However, the PGAA technique has been applied to a wide variety of situations, ranging from in vivo analysis¹⁵ to precise determination of contaminants in new complex materials16 worldwide. A compiled list of reactor based PGAA facilities are given in Table 2. The Table 2 lists years of first PGAA report, reactor power level, neutron beam type, neutron beam size, neutron flux, cadmium ratio, detector to sample distance, detection system, and measured sensitivity for boron, for each facility. Table 3 is a list of references for each facility provided. An IAEA Technical Document on ìDatabase of Prompt Gamma Rays from Slow Neutron Capture for Elemental Analysis" converses further characteristics of modern PGAA facilities and available through chouse@iaea.org.¹⁷

The applications of the PGAA facilities include: (1) determination of B and Gd in biological samples which are used for neutron capture therapy (NCT) studies, (2) determination of B, and H impurity levels in metals, alloys, and semiconductors, (3) multielemental analysis of geological, archaeological, and environmental samples for determinations of major components such as Al, S, K, Ca, Ti, and Fe, and minor or trace elements such as H, B, Cl, V, Mn, Co, Cd, Nd, Sm, and Gd, and (4) multielemental analysis of biological samples for the major and minor elements H, C, N, Na, P, S, Cl, and K, and trace elements like B and Cd. The analysis of biological materials has been one of the main concerns of the PGAA development.

Through the use of cold neutrons, curved neutron guides, and neutron focusing systems, modern PGAA facilities has greatly improved detection sensitivities, comparable to instrumental neutron activation analysis, allowing PGAA to be applied to many technical problems involving analytical chemistry.

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