LA-UR-96- 3225

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Efficiency**

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Development of a High-Count-Rate Neutron Detector with Position Sensitivity and High Efficiency

Ronald Nelson* and John Sandoval

Abstract

This is the final report of a one-year, Laboratory-Directed Research and Development (LDRD) project at the Los Alamos National Laboratory (LANL). While the neutron scattering community is bombarded with hints of new technologies that may deliver detectors with high-count-rate capability, high efficiency, gamma-ray insensitivity, and high resolution across large areas, only the time-tested, gas-filled ^3He and scintillation detectors are in widespread use. Future spallation sources with higher fluxes simply must exploit some of the advanced detector schemes that are as yet unproved as production systems. Technologies indicating promise as neutron detectors include pixel arrays of amorphous silicon, silicon microstrips, microstrips with gas, and new scintillation materials. This project sought to study the competing neutron detector technologies and determine which or what combination will lead to a production detector system well suited for use at a high-intensity neutron scattering source.

1. Background and Research Objectives

While the neutron scattering community is bombarded with hints of new technologies that may deliver detectors with high-count-rate capability, high efficiency, gamma-ray insensitivity, and high resolution across large areas, only the time-tested, gas-filled ^3He and scintillation detectors are in widespread use. Such detectors are used almost exclusively at the Los Alamos Neutron Science Center (LANSCE), but with our current neutron fluxes these technologies impose count-rate limitations. This project sought to identify which advanced detector schemes that are as yet unproved in production systems would best work with future spallation sources with fluxes a factor of 10 to 100 greater than currently available at LANSCE.

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The international community is developing new gas, scintillating, and solid-state detectors for neutron detection. One promising evolution involves microstrip gas detectors pioneered at the Institute Laue Langevin (ILL) [1]. Using a photolithographed anode/cathode-type detector deposited onto glass, this detector has achieved 78% efficiency for 1.2-Å neutrons with a gas mixture of 3-bars ^3He and 1-bar CF_4 , has a resolution of 200 μm , and has maximum count rates of 2.0×10^6 per strip. Commercial vendors are also addressing the needs of the community by improving performance of ^3He -detector response times of 50 ns rise time and less than 3 μs fall time [2].

Since photons from scintillators can be collected rapidly compared with charge collection in traditional gas-tube detectors, ^6Li scintillation detectors can, in principal, overcome the count-rate limitation. While scintillators are also sensitive to gamma radiation, gamma sensitivity can be reduced with pulse shape discrimination by two orders of magnitude, if one is prepared to suffer a 50% reduction in neutron efficiency. The Rutherford Appleton Laboratory has pioneered the development of gamma-insensitive ZnS scintillation detectors for use at its ISIS facility.

One new solid-state, thermal-neutron detector [3] showing tremendous promise for near future applications utilizes a multi-layer configuration of a hydrogenated amorphous silicon, a-Si:H. Despite its poorer electronic characteristics, compared to crystalline silicon, a-Si:H offers strong advantages of availability in large dimensions and much higher radiation resistance. This material has been successfully used as a detector for X-rays, gamma rays, charged particles and neutrons [4,5].

Gas and scintillation detectors are currently in use at LANSCE for neutron detection and both types have count-rate limitations that make them inadequate for the high cold- and thermal-neutron fluxes anticipated for LANSCE II. Table 1 summarizes the anticipated peak and time-averaged count rates for some of the instruments proposed for LANSCE II, an upgrade of the current LANSCE facility. The inadequacy of present technology in dealing with these rates is demonstrated by comparison with some present LANSCE instruments. At LANSCE the Low Q Diffractometer (LQD) is saturated with rates of 30 KHz and the High-Intensity Powder Diffractometer (HIPD) has neutron pileup events at rates of 100 KHz per detector. The standard counting ^3He detectors are quoted as having maximum rates of 10^5 neutrons per second [6,7]. Commercial area detectors are quoted with maximum rates of 10^5 and typical efficiencies of 52% for 1 Å [8]. Linear position-sensitive detectors exhibiting resolution of 3 mm have been reported with efficiencies of 64% at 1 Å and 74% at 1.3 Å; however, the maximum rates are only 5×10^3 counts per second [9]. The need for high-efficiency detectors capable of high count rates is already being felt at LANSCE since the current gas ^3He detectors are not adequate to process data at the 106-Hz level.

Table 1. Count rates for LANSCE II Instrumentation

<u>Diffractometer Instruments</u>	<u>Count rate information</u>
(1) Powder Diffractometers:	
(a) NPD	measurement times ≤ 30 min.
(b) ENG I & II	measurement time $\geq 50 \mu\text{s}$
(c) Single Shot	$1-2 \times 10^6$ n/s total, 600-1,600 / detector tube
(d) HIPD	(1-2 min measuring times.)
(e) Special Environment/ LASD	
(f) HRPD	
(g) Magnetism Powder Diffractometer	
(2) Single-Crystal Diffractometers:	
(a) chemical crystallography	4, 2D sectors 1 MHz/ sector, 4 MHz overall
(b) protein crystallography	4, 2D sectors 1 MHz/ sector, 4 MHz overall
(c) Structural Biology X station	
(d) diffraction physics (polarized beam)	
(3) Low-Q Diffractometers:	
(a) MQD (with wide angle detectors)	1 MHz over entire 2D detector peak, 200 kHz time average
(b) LQD (with polarized beam capability)	200 kHz peak, 50 kHz time average
(c) Biological LQD	100 kHz peak, 25 kHz time average
(d) VLQD	10 kHz peak, 2.5 kHz time average
(4) Reflection Geometry Diffractometers:	
(a) Neutron Reflectometer	
(b) Polarized-Beam Neutron Reflectometer	
(c) High-Intensity Neutron Reflectometer	
(d) Grazing Incidence Neutron Diffractom.	

2. Importance to LANL's Science and Technology Base and National R&D Needs

This project supports Los Alamos core competencies in complex experimentation and measurement as well as nuclear science, plasmas, and beams. It also provides necessary data for upgrading the detection equipment at LANSCE.

3. Scientific Approach and Results to Date

Our research focused on the literature and on contacts with staff at other laboratories both in the United States and Europe. From a field of technology options including micro-strip gas detectors, linear position-sensitive detectors, scintillation detectors, and solid-state area detectors, we identified the two technologies that offer the maximum benefit for projected LANSCE research, gas micro-strip, and solid-state area detectors.

Micro-strip Gas Detectors

Despite their disadvantages ^3He gas detectors are still evolving; the proportional gas detector pioneered by Charpak and Sauli [10] is now the focus of renewed attention. The major difference between older multiwire proportional counters and the new microstrip types are that the wires are now replaced with a microstrip that is etched or deposited on either a glass substrate or a silicon substrate. Submillimeter resolution is possible with microstrip detectors. The pattern for the glass substrate can be designed by the user and a photolithographic technique is used to deposit the pattern on a sheet of glass [1]. If a silicon substrate is used, the size of the wafers, four-inch standard, and the deposition process uses more costly etching techniques. However, if silicon wafers are used an amplifier could be deposited beneath the strip. This process could significantly reduce the cost of instrumenting a detector system.

At the ILL, A. Oed is building the "banana detector," a four-foot, banana-shaped detector [11]. The detector has a photolithographed anode/cathode-type detector deposited onto Schott #S8900 glass. The glass is selected for its resistive nature (10^{13} - 10^{14} ohms) and must be devoid of pits or pin-holes. The gas is at four-bars pressure and consists of 3-bars ^3He and 1-bar CF_4 . The pressure results in 79% efficiency for 1.2-Å neutrons. Cleanliness is stressed with the chamber constructed of stainless steel and ceramic. If any glues, adhesives or solder resin reside in the chamber, the lifetime of the chamber decreases. The CF_4 reacts with the contaminants to form higher order compounds that attach to the strips causing breakdown or leakage currents (noise, dark current). Recently a microstrip-prototype-detector test showed the detector lost efficiency due to neutron radiation damage [12]. Despite this early result, the ILL believes that an appropriate glass will solve the observed efficiency degradation.

Linear Position-Sensitive Detectors

Linear position-sensitive detectors (PSDs) can be thought of as inexpensive building blocks for an area detector. The observed area has much greater throughput than a ^3He gas area detector. Typically, a single element (linear PSD) is tied to a dedicated output channel and although the rates could be 10^4 - 10^5 , the overall rate for the detection area is calculated as n-elements times the individual rate. Ordela, Inc., is proposing a "mosaic" detector that is a stack of linear PSDs used as an area detector. The overall output from the detector is increased because each linear PSD has a separate readout. Ordela, Inc., is collaborating with the University of Tennessee and Oak Ridge National Laboratory (ORNL) to produce the area detector. The University of Tennessee is developing application specific integrated circuits (ASIC) and multi-chip modules (MCMs) for the data acquisition.

Scintillation Detectors

Typical scintillator density is roughly two orders of magnitude greater than the densities of gases in proportional counters. This means the scintillator can be thin and absorption efficiency can be high for slow neutrons. However, the density also ensures that the scintillator material will absorb energy from fast neutrons and gamma rays. By selecting an optimum geometry and electronic discrimination, slow neutron detection can be optimized. These types of detectors are typically developed at the neutron scattering institution [13] and are not commercially available. Although neutron-sensitive glass and plastic are available commercially [14,15] and a single slow-neutron detector (Si diode type) is available [16], the only commercial ventures for slow-neutron area detector systems (scintillation type) are research proposals. One small-business, innovative-research (SBIR) proposal demonstrates feasibility for a ZnS sheet coupled to a photodiode array [17]. The Phase II proposal would provide a prototype two-dimensional detector (20 cm x 20 cm with 1-mm resolution) and the Phase III would provide a 1-m x 1-m array with 1-mm resolution [18]. Another proposal describes a ^6Li -glass (7 mm x 7 mm) coupled array where each pixel (5 mm) is coupled to a signal-processing ASIC, data-logic ASIC, and a storage array. The intent is to build a scaleable detector made of 7-cm x 7-cm area segments [19].

Another unique solution presented by Miley [20] proposed using three bundles of (neutron-sensitive) fiber-optic cable for detection. The three bundles are used in an "exclusive or" condition for thermal neutron detection and in an "and" condition for the detection of gamma rays. A single fiber provides approximately 20% efficiency and a bundle of six fibers provides 95% efficiency. This fiber is under patent to Battelle [21] and is used in portal monitors for detection of special nuclear materials. The scintillating fibers are coupled to a photomultiplier tube (PMT) for amplification and conversion of the light output. In the past, PMTs have been rather large and require power supplies capable of supplying voltages of 2 kilovolts. Recently, HamamatsuTM announced a PMT housed in a TO-8 metal can (10 mm in length and 15 mm in diameter) [22]. This PMT could be used in an area detector and provide the needed high rate, and because of its physically small size, added resolution. This detector scheme requires intensive manpower initially for construction and calibration, and in addition the scintillating glass or fiber as well as the photomultiplier tube and base are costly, but long-term maintenance is minimal. Because of the low maintenance requirements the scintillation type detectors are very attractive for slow-neutron detection.

The advantages of scintillation detectors are high count rate, low maintenance, and long lifetime. The disadvantages of scintillation detectors are gamma-ray sensitivity, low effective efficiency for slow neutrons, and high initial expense for glass, photomultiplier tubes, and skilled manpower to assemble the detector. Recent work for scintillators is summarized in Table 2.

Table 2. Area Scintillation Detectors.

<u>Institution</u>	<u>Material</u>	<u>Amplifier</u>	<u>Rate</u>
ORNL [23]	Li	PMT	2 MHz
RAL [24]	ZnS/ Li	PMT	400 kHz
Nanoptics, Inc. [25] (Detector only)	Plastic $^6\text{Li}(^{10}\text{B}$ enriched)	NA	NA--SBIR Phase 1 completed

Note: The Nanoptics entry is for a new neutron capture plastic only, not a detection system.

Although solid-state detectors are doped with lithium and boron, the concentrations are not sufficient for high absorption efficiency. Typically, solid-state detectors must rely on converters for neutron capture. Neutron-capture efficiencies are low because of the charged-particle path length. Therefore, solid-state detectors are still waiting for an appropriate material that possess high-capture efficiencies and still maintain useful semiconductor properties. Foil detectors use a coating of neutron-sensitive material coupled to a solid-state device for neutron detection. However, a significant trade-off exists between foil thickness and capture efficiency. Capture efficiencies can be increased by creating a multilayered device, but scattering increases and results in lower signal levels.

Solid-state-type detectors hold the most promise for future detection systems. These types of detectors do not require exotic gases and kilovolt power supplies. They can be produced in quantity with submillimeter resolutions. One of these new thermal neutron detectors using gadolinium and amorphous silicon has been tested by Miresghhi, *et al.*, [26]. The detector uses two layers of gadolinium to provide a thermal-neutron-detection efficiency of 42%. The gamma-ray sensitivity is approximately 1×10^{-5} . Miresghhi further claims that an enriched-Gd layer could boost efficiencies to 63% [27]. It is important to note that although rapid progress has been made on the detector, much remains to be studied. For example, an array of detectors has not been designed or tested and the noise/coupling between pixels has not been characterized. Of the three types of detectors for thermal-neutron capture the silicon-

gadolinium detectors with embedded readout and amplifier would be the simplest to operate and require the least maintenance. Table 3 lists the most recent amorphous-silicon, solid-state detectors, and the converter for the device. A recent research effort at Grumman has produced a portable detector for environmental assay. Although the detector has low efficiency (13%), the manufacturer claims that the detector was made with natural gadolinium (supplied by ORNL) and coupled to an eight-channel commercial charge preamplifier and that no special processes were involved in the development of the detector system.

Table 3. Solid State Area Detectors

<u>Institution</u>	<u>Converter</u>	<u>Technology</u>	<u>Rate</u>
Grumman [28]	Gadolinium Foil	a-Si-diode	62 kHz
LBL [27]	Gadolinium Foil	a-Si -Thin Film Trans.	NA
Advanced Optical Technologies [29]	ZnS	a-Si-photodiode	NA--SBIR phase 1 completed

Detector development for LANSCE II is crucial for the effective utilization of the neutron count rates that will be available from the 330- and 660-kW targets. Because the need is immediate and critical we will proceed in two stages. The first is to design and build multiwire area detectors with multichannel amplification. This should increase the maximum peak count rates for position-sensitive ^3He multiwire proportional counters to the 1-MHz range. After development of a version intended for the protein crystallography and biological crystallography stations, others suitably modified can be made for small-angle and neutron-reflectometry applications. Parallel with the development of fast gas detectors, work on solid-state semiconductors will be undertaken, though clearly the time needed to realize these detectors will be longer than the development of gas detectors. Ultimately, it is envisioned that solid-state detectors will have better performance, particularly with respect to the highly critical area of peak count-rate capability.

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