Update on a ZnO:Ga Alpha Particle Detector for a Portable Neutron Generator for the Nuclear Materials Identification System (NMIS)

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ABSTRACT

We report results from efforts to develop a recoil alpha particle detector for use in a portable neutron generator. The associated particle sealed tube neutron generator (APSTNG) will be used as an interrogation source for the Nuclear Materials Identification System (NMIS). With the emission of 14 MeV neutrons produced by the D-T reaction, associated 3.5 MeV alpha particles are emitted. These neutrons and alphas may then be correlated in time and position, thus effectively "tagging" the neutrons of interest for subsequent use as an active nuclear materials interrogation source. The alpha particle detector uses a ZnO:Ga scintillator coating applied to a fiber optic face plate. Gallium-doped zinc oxide is a fast (decay time < 1ns), inorganic scintillator with a high melting point (1975ºC). The scintillator is coated with a thin layer of aluminum in order to screen out light produced in the tube and scattered deuterons. This coating also serves to prevent the buildup of charge on the detector surface. Previous measurements used small sample sizes for coating thickness optimization studies. Our most recent measurements were made with full size prototype fiber optic face plates. These measurements have provided confidence to fabricate a detector that is ready for installation in an APSTNG.

INTRODUCTION

The Nuclear Materials Identification System (NMIS) time-dependent coincidence processor has been described in a previous work [1]. Primarily used for identification and characterization of highly enriched uranium (HEU) and plutonium, the present NMIS configuration consists of 1GHz synchronous sampling of five channels, associated software, advanced data analysis methods, and an instrumented (ion chamber) Cf-252 fission source. Incorporation of a small, lightweight, portable deuterium-tritium (DT) neutron generator as an active interrogation source will provide several improvements for the NMIS. These improvements include high energy (14.1 MeV), nearly monoenergetic neutrons, single neutron emitted per source event, the ability to turn off the source of neutrons, and higher sensitivity.

The associated particle technique "tags" emitted 14.1 MeV neutrons through detection of the associated 3.5 MeV alpha particle produced by the D-T reaction. This directional tagging of the neutrons, coupled with time tagging by the NMIS, will reduce background sources in the NMIS signatures and increase sensitivity. This work reports efforts to develop a scintillator based alpha particle detector that will define a cone of tagged neutrons. This report focuses on efforts to test three full-size prototype alpha particle detectors.

ASSOCIATED PARTICLE SEALED TUBE NEUTRON GENERATOR

To incorporate an APSTNG into the NMIS, we considered the following requirements:

- Must be portable (approximately 40 inches long, 3 inches in diameter, weight less than 30 pounds)
- Must be capable of producing a total neutron flux of 10^8 neutrons per second during routine operation
- Must be of a sealed-tube design to reduce risk of tritium contamination to environment

The APSTNG for Oak Ridge National Laboratory (ORNL) is being built by Thermo Electron Corporation of Colorado Springs, Colorado. The alpha particle detector will be developed and manufactured by ORNL, in conjunction with Purdue University, and installed by Thermo Electron. The ion source reservoir and target are designed such that, after several hours of operation, both contain deuterium and tritium in equilibrium. This allows stable neutrons yields over the life of the generator. Figure 1 presents a representative schematic of the ORNL APSTNG.

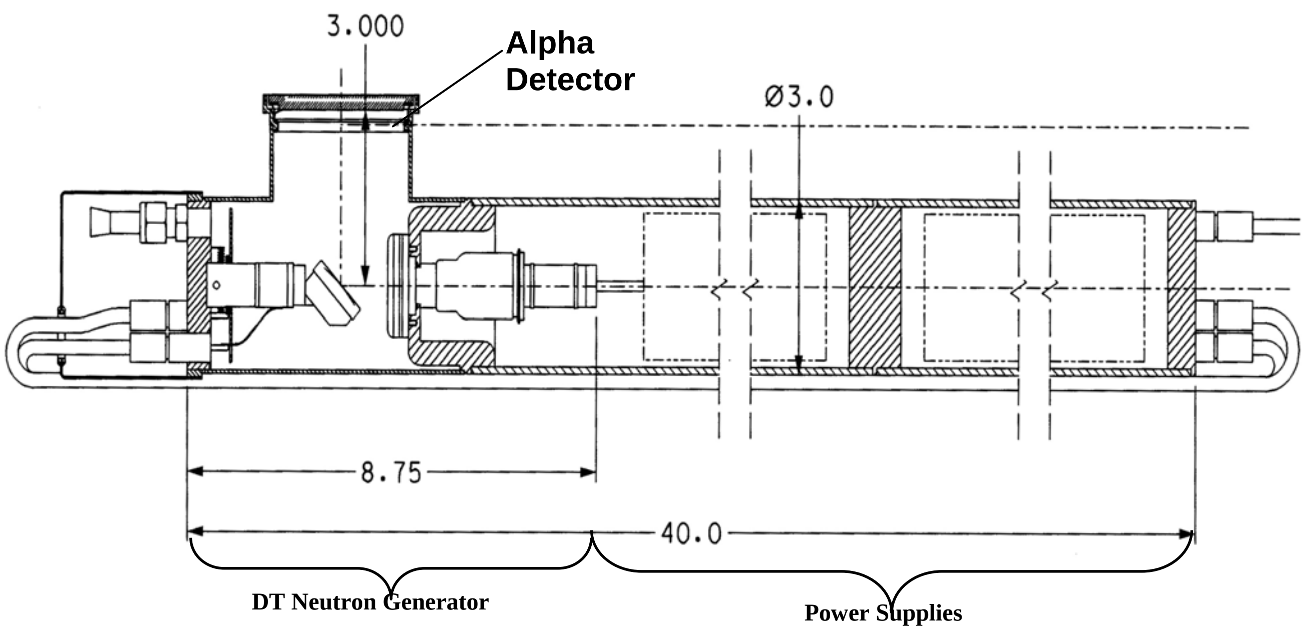

Figure 1. ORNL APSTNG representative schematic.

The alpha detector will be placed in a three inch diameter port that is mounted at a 45° angle from the target face. Detected alpha particles define a cone of oppositely directed neutrons, thus effectively direction tagging the neutrons. Initial efforts will focus on defining this cone of neutrons with a diaphragm iris placed between the transducer and the photomultiplier tube (PMT) while future efforts will consider pixelating the alpha detector for finer position resolution.

ALPHA PARTICLE DETECTOR

This effort considered a scintillator as the alpha particle transducer and is based on the work of Beyerle, Hurley, and Tunnell [2]. A parallel effort within our group is pursuing an alternative design that utilizes a segmented silicon diode detector. Several design specifications were considered for construction of the alpha transducer. These specifications included:

- Fast (ns regime) timing
- Ability to survive bakeout at 350°C over several days
- No organics used for phosphor coating
- Attempt to maintain position resolution
- Thin film metal overcoating to reduce background from scattered deuterons and tritons

ZnO:Ga had been previously considered [3] and utilized [4] as a phosphor coating for an APSTNG alpha transducer. This inorganic phosphor has a fast decay time of < 1 ns, a high melting point of 1975°C, and a light yield up to ~3300 photons per MeV of deposited energy.

Previous measurements used small sample sizes for phosphor coating thickness and metal overcoating optimization studies [5]. One-inch round planar fiber optic face plates and one-inch round fused silica discs served as the substrates for the transducers. The face plates incorporated ~5% "black" fibers among the transmitting fibers in order to reduce cross-talk amongst neighboring fibers. Measurements indicated an optimum phosphor coating thickness in the range of 2-4 mg/cm^2. Light output measurements for 5 mg/cm^2 samples indicated significant light loss due to self-attenuation. Fiber optic face plates reduced light output by 15-20% but produced more uniform light output over the range of coating thicknesses. Previous calculations and measurements examined the optimization of the thin film metal overcoating. The metal coating was applied to serve several functions:

- to stop deuterium and tritium ions scattered from the ion beam and the target
- to stop low energy alpha particles that are produced at relatively great depths in the target,
- to act as a light barrier thus protecting the coupled PMT
- to prevent charge buildup on the underlying window.

Careful consideration has been given to the application of this thin metal film since in one case, an inadequate film thickness led to high count rates in the alpha detector due to scattered deuterons [3]. Although the previous study indicated nickel to be a better choice than aluminum as a metal overcoating due to aluminum adhesion problems, the full size prototype face plates used an aluminum overcoat. No further aluminum adhesion problems have been observed.

Given the results from the previous work using small sample sizes, three full size prototype detectors were fabricated using ~3-inch fiber optic face plates. Figure 2 shows a photograph of one of these prototype detectors with its surrounding metal flange. Two of the face plates incorporated ~5% "black" fibers among the transmitting fibers in order to reduce cross-talk among neighboring fibers. ZnO:Ga was applied by Lexel Imaging, Inc. [6] using a gravity settling technique that used potassium silicate as the binder and a proprietary compound as the electrolyte. Phosphor coatings were applied in thicknesses of ~3 mg/cm^2 which were subsequently overcoated with ~1.5 microns of aluminum. The two detectors designated A and B include black fibers, while detector C does not. Detector B has been successfully installed in an APSTNG and detector C has been recently fabricated and undergone initial testing.

Figure 2. Photograph of prototype ZnO:Ga and aluminum coated fiber optic face plate with attached flange.

TEST RESULTS

The test setup consisted of a Burle 8850 Quantacon photomultiplier tube (PMT) and a pulse height spectroscopy system (preamplifier, shaping amplifier, and multichannel analyzer (MCA)). Tests were made at various alpha particle energies, from 1.8 MeV to 5.2 MeV, using varying foil thicknesses. Alpha particles were collimated using a 0.125-inch aperture. A silicon surface barrier detector was used for comparing measurements between the ZnO:Ga and silicon detectors and calculating counting efficiencies. A blue light emitting diode (LED) and a R4885 PMT were used for light leak checks.

A typical mapping pattern for light output measurements and efficiency calculations is shown in figure 3. Background runs were made with 10 mm of Aluminum inserted between the Am-241 source and the coating. The light output was evaluated by calculating the first moment of the spectrum, after background subtraction. Figures 4 and 5 show light output as a function of incident alpha energy for coatings B and C. The light is provided in photoelectrons following a light output calibration using a pulsed blue LED.

The peak of the ZnO(Ga) emission spectrum is at 390 nm [7]. In this wavelength range, the quantum efficiency of the 8850 PMT is 20-25%, so a light output of 70 pe on average corresponds to roughly 300 photons impinging on the photocathode per alpha event.

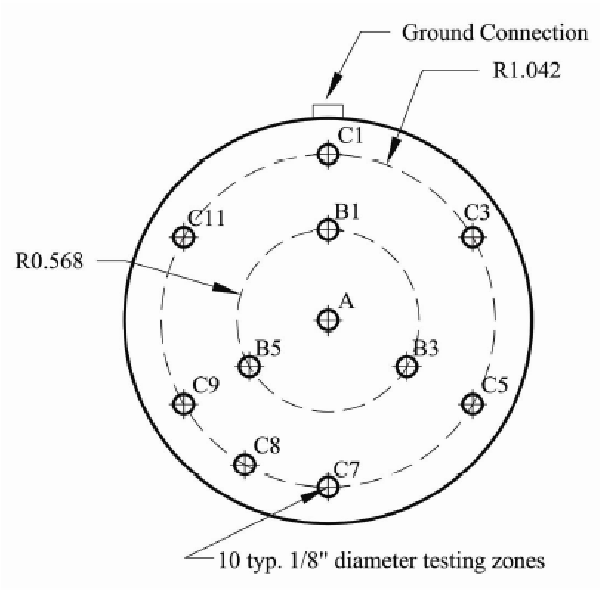

Figure 3. Typical mapping pattern for light output measurements and efficiency calculations.

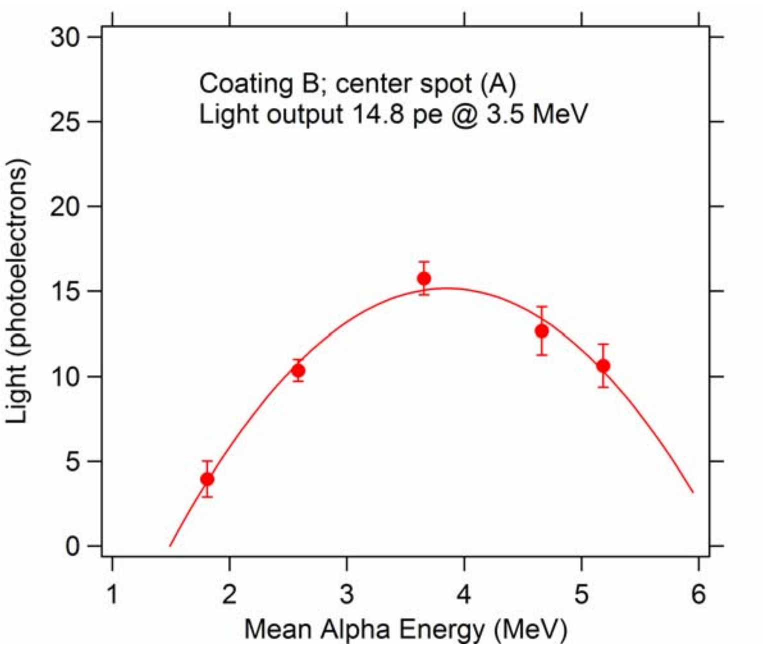

Figure 4. Calculated light output for coating B. Light output is provided in photoelectrons following a calibration of the PMT using a pulsed blue LED.

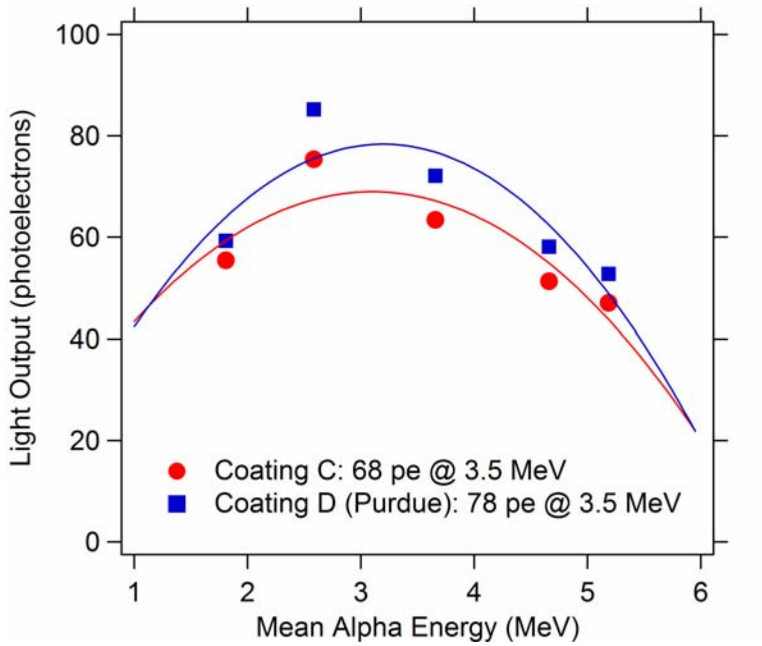

Figure 5. Calculated light output for coating C. Light output is compared to a similarly fabricated face plate made for Purdue University.

Efficiency measurements and calculations were made using the Quantacon PMT and the silicon surface barrier detector. Data were taken at the same energies as for the light output measurements and background subtracted. Quoted efficiencies are relative to the silicon detector. Figure 6 shows efficiency versus incident alpha particle energy for coating C with an average efficiency of 94%, not including the 1.8 MeV measurement.

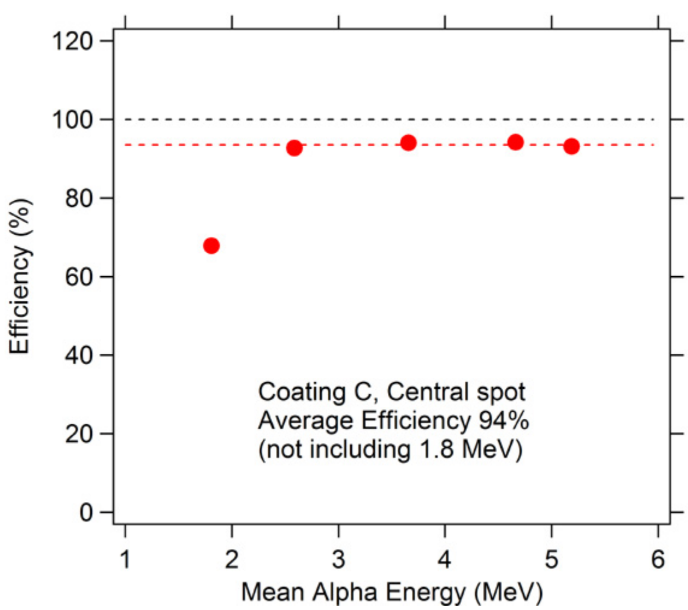

Figure 6. Calculated efficiency versus incident alpha particle energy for coating C.

One of the functions of the metal overcoat is to act as a shield against light produced in the APSTNG. While the light generation is not well characterized for our APSTNG and communications with users of similar APSTNGs indicate light generation is not a significant problem, we believed it was prudent to characterize our coating's performance. The face plate was attached to the face of a 3-inch R4885 PMT with dark tape up to the front edge of the plate such that light could not enter from the side of the plate. The coating was illuminated by a blue LED in short bursts. The plate was removed and the PMT was illuminated by the same LED in the same position. Without the plate in place, the PMT was operated at significantly lower gain (factor of ~6380). It was determined that the coating attenuates light from a blue LED by a factor of at least 400,000. Figure 7 shows the results of the light leak measurements for coating B.

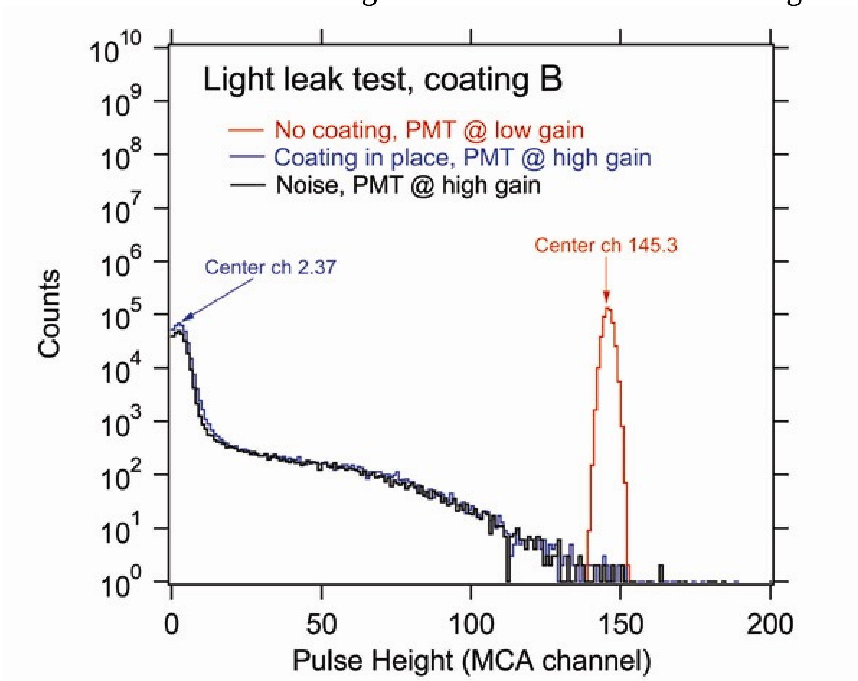

Figure 7. Light leak measurements for coating B.

SUMMARY

We have developed and tested full sized ZnO:Ga coated fiber optic face plates in efforts to develop an alpha particle detector for an APSTNG. Results to date indicate promise as an effective alpha particle detector for the APSTNG for future use in the NMIS as an active interrogation source allowing direction tagging of 14.1 MeV neutrons. While the light output per event is low, it is distinguishable above background, especially for face plate C that does not use cross-talk reducing black fibers. These measurements have provided confidence to fabricate a detector that is ready for installation in an APSTNG. Recently, detector B has been successfully installed in an APSTNG that is ready for testing.

ACKNOWLEDGEMENTS
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