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Evaluation of ZnO(Ga) Coatings as Alpha Particle Transducers Within a Neutron Generator

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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 investigations and preliminary 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 14.1 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 direction, 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 (1.5 ns decay time), inorganic scintillator with a high melting point (1975C) and an absolute light yield of 1.5% of NaI(Tl). The scintillator is coated with a thin layer of nickel in order to screen out light produced in the tube and scattered deuterons and tritons. This coating also serves to prevent the buildup of charge on the detector surface. Results to date indicate promise as an effective alpha particle detector for the APSTNG for future use in the NMIS.

INTRODUCTION

The Nuclear Materials Identification System (NMIS) time-dependent coincidence processor has been thoroughly described in 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 may provide several improvements for the NMIS. These improvements include

- high energy (14.1 MeV), nearly monoenergetic neutrons
- single neutron emitted per source event
- ability to turn off the source of neutrons
- 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 direction tagging of the neutrons, coupled with time tagging by the NMIS, will reduce background sources in the NMIS signatures. 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 develop the alpha particle transducer portion of the alpha detection system.

Potential applications of this combination DT generator and alpha particle detector include, but are not limited to:

- Arms control treaty verification
- Nuclear material control and accountability
- Mine detection
- Detection of explosives, chemical warfare agents, illicit drugs

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 10^7 14.1 MeV 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) will be built by MF Physics Corporation of Colorado Springs, Colorado[2]. The alpha particle detector will be developed and manufactured by ORNL and installed by MF Physics. 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. Operating currents up to 100 microamps are possible. Figure 1 presents a representative schematic of the ORNL APSTNG.

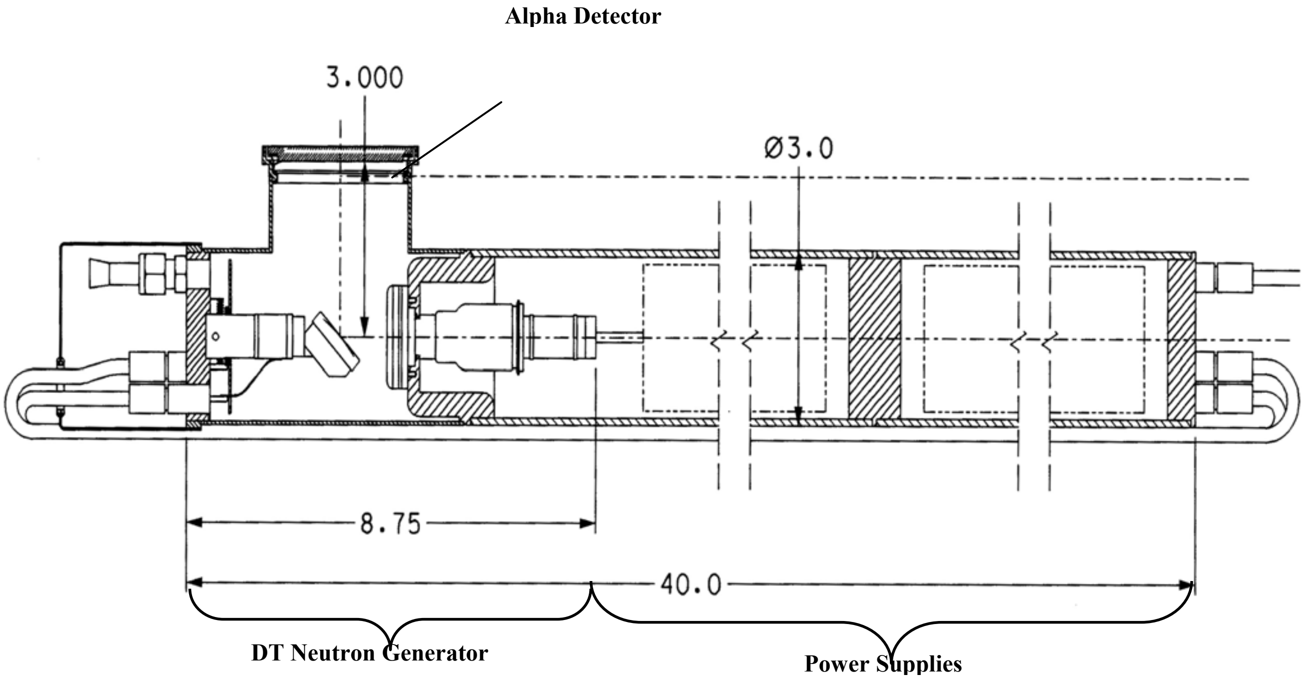

Figure 1. ORNL APSTNG representative schematic.

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 are focused 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 TRANSDUCER

This effort considered a scintillator as the alpha particle transducer and is based on the previous work of Beyerle, Hurley, and Tunnell [3]. 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 300°C over several days
- No organics used for phosphor coating
- Attempt to maintain position resolution
- Thin film metal coating to reduce background from scattered deuterons and tritons

ZnO(Ga) had been previously considered [4] and utilized [5] as a phosphor coating for an APSTNG alpha transducer. This inorganic phosphor has a fast decay time of ~1.5 ns, a high melting point of 1975°C, and a light yield that is about 1.5% of NaI(Tl), thus producing ~560 photons per MeV of deposited energy.

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 incorporates ~5% “black” fibers among the transmitting fibers in order to reduce cross-talk amongst neighboring fibers. Fiber optic face plates are being considered for future work to pixelate the alpha detector.

The phosphor coating was deposited on fiber optic face plates and fused silica disks for testing. ZnO(Ga) was applied by Lexel Imaging, Inc. [6] using a gravity settling technique that used potassium silicate as the binder and strontrium nitrate as the electrolyte. Coatings were applied in thicknesses of 2, 4, 5 mg/cm^2. Calculations for 3.0 MeV alphas (alphas lose about 0.5 MeV in passing through the nickel overcoating discussed below) normally incident on ZnO(Ga) were made using the Monte-Carlo Transport of Ions in Matter (TRIM) program [7], one of a group of programs from the Stopping and Range of Ions in Matter (SRIM) program package. The calculated range was about 7 microns. The model assumed a ZnO(Ga) crystal density of 5.6 g/cm^3, the theoretical density for ZnO(Ga) and a one percent stoichiometric gallium composition . While the packing density of individual crystals in the binder is not known, these calculations provided a reasonable starting point for specifying coating thicknesses.

Previous work [4,5] to produce an alpha detector considered the application of a thin film (1-2 µm) of aluminum or nickel on top of the ZnO(Ga) phosphor. This 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 [4]. Calculations for 75 keV deuterons and tritons normally incident on aluminum and nickel were performed using the TRIM program. Mean projected range and longitudinal straggling, defined as the second moment of the distribution, results for 10^4 incident ion histories are presented in Table 1.

Ion/Coating	Mean Projected Range (μm)	Longitudinal Straggling(μm)
Deuteron/Aluminum	0.7538	0.1007
Deuteron/Nickel	0.4136	0.0955
Triton/Aluminum	0.8670	0.1209
Triton/Nickel	0.4753	0.1149

Table 1. Calculated ranges for deuterons and tritons incident on aluminum and nickel.

Nickel was chosen over aluminum as the thin film metal coating as there were concerns about the adhesion of the metal coating to the underlying phosphor and window. A thinner metal coating was thought to be less likely to peel away from the window. An added benefit of using nickel was the reduced neutron activation of the nickel (five neutron absorptions required to activate Ni-58) as compared to aluminum (1 neutron absorption required to activate Al-27).

One micron thick nickel coatings were applied to each of three phosphor-coated silica disks with phosphor thicknesses of 2, 4, and 5 mg/cm^2 using an electron beam evaporation technique. The phosphor-coated fused silica disks were first heated to 150°C in order to drive off water vapor as well as to aid in nickel adhesion to the windows. Bakeout tests at 300°C for four days have demonstrated the satisfactory adhesion of nickel to the underlying phosphor.

TEST RESULTS

Testing of our phosphor-coated silica disks, with and without nickel coatings, and fiber optic face plates, without nickel coatings, has been previously reported [8]. Samples were tested with 5.5 MeV alpha particles from an Am-241 source. A reference sample of BC-400 scintillating plastic was also tested. Light output, in units of photoelectrons, was measured using a Burle 8850 Quantacon PMT. The PMT output signal was split into a LeCroy WavePRO 960 digital sampling oscilloscope and LeCroy 825 Risetime-corrected Discriminator. Dual discriminator outputs were used to trigger the oscilloscope and to drive a Joerger VS scaler. Tests were made at various alpha particle energies (using varying spacings of intervening air between the source and the coatings) for the different coating thicknesses. Table 2 presents a summary of measured light outputs.

Substrate/ Overcoat	Coating Weight (mg/cm^2)	Light Output @ 3.2 MeV	Light Output @ 5.4 MeV
Silica	2	59.0	55.9
Silica	4	53.9	61.0
Silica	5	20.1	35.9
Fiber Optic	2	46.8	47.6
Fiber Optic	4	45.8	49.2
Fiber Optic	5	-	45.2
Silica/Ni	2	44.4 (@3.5 MeV)	32.5
Silica/Ni	4	35.5 (@3.5 MeV)	49.9
Silica/Ni	5	-	20.4
BC-400	-	114.8	200.1

Table 2. Summary of light output results for various transducer substrates and phosphor coatings.

Results from Table 2 indicate an optimum phosphor coating thickness in the range of 2-4 mg/cm^2. Light output measurements for 5 mg/cm^2 samples indicate 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. Figure 2 presents a representative pulse height spectrum for a silica disc coated with 4 mg/cm^2 of phosphor excited by 3.2 MeV alphas.

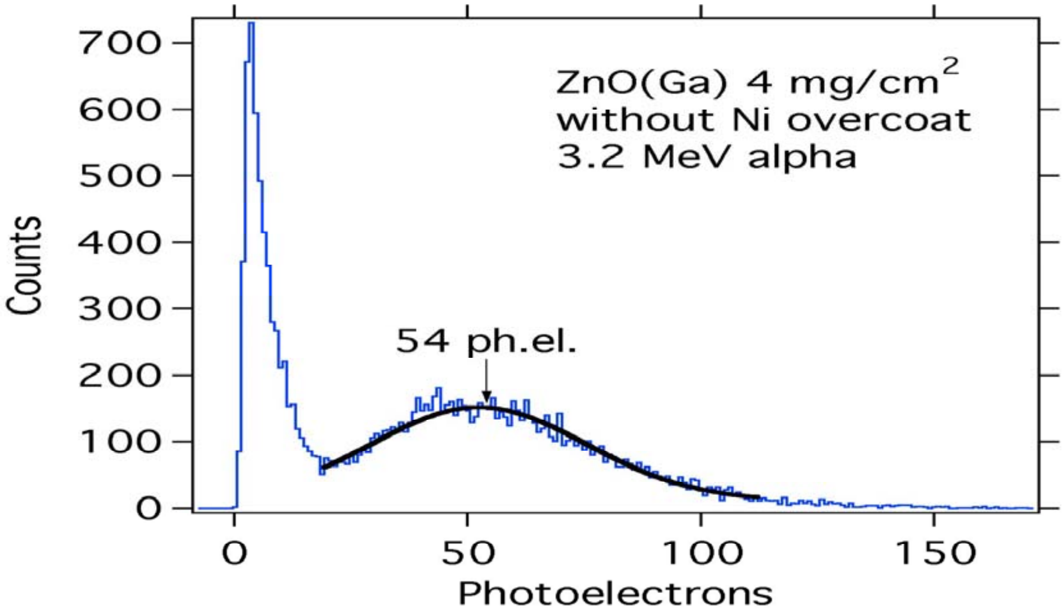

Figure 2. Pulse height spectrum for silica disc coated with 4 mg/cm^2.

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FUTURE WORK

Future measurements include light output tests after bakeout of the samples, light output tests of nickel-coated fiber optic face plate samples, and testing of larger (68 mm diameter), concave fiber optic face plate prototypes. Detector efficiency measurements and calculations for all samples are ongoing.

SUMMARY

We have developed and tested various ZnO(Ga) coated substrates 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. Direction tagging will reduce background sources in NMIS signatures.

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