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FIGARO: Detecting Nuclear Material using High-Energy Gamma Rays from Oxygen

B. J. Micklich[a], D. L. Smith[a], T. N. Massey[b], D. Ingram[b], and A. Fessler[a]

[a]*Technology Development Division, Argonne National Laboratory, Argonne, IL 60439 USA*
[b] *John E. Edwards Accelerator Laboratory, Ohio University, Athens, OH 45701 USA*

Abstract. Potential diversion of nuclear materials is a major international concern. Fissile (e.g., U, Pu) and other nuclear materials (e.g., D, Be) can be detected using 6-7 MeV gamma rays produced in the $^{19}F(p,\alpha\gamma)^{16}O$ reaction. These gamma rays will induce neutron emission via the photoneutron and photofission processes in nuclear materials. However, they are not energetic enough to generate significant numbers of neutrons from most common benign materials, thereby reducing the false alarm rate. Neutrons are counted using an array of BF_3 counters in a polyethylene moderator. Experiments have shown a strong increase in neutron count rates for depleted uranium, Be, D_2O, and ^6Li, and little or no increase for other materials (e.g., H_2O, SS, Cu, Al, C, ^7Li). Gamma source measurements using solid targets of CaF_2 and MgF_2 and a SF_6 gas target show that proton accelerators of 3 MeV and 10-100 microamperes average current could lead to acceptable detection sensitivity.

INTRODUCTION

Significant quantities of special nuclear material (SNM) have been smuggled across borders or facility boundaries. This poses a major national and international concern. The danger could be reduced substantially by developing an effective, compact, transportable, and affordable inspection system that could be deployed easily at critical locations, e.g., border crossings or sensitive nuclear facilities.

The FIGARO concept uses a compact, low-energy proton accelerator to generate gamma rays that can interrogate objects such as packages, luggage, or containers for the presence of all SNM, including U-235, as well as other nuclear materials such as beryllium and deuterium. FIGARO gives an unambiguous signal for the presence of nuclear material, independent of size, shape, or chemical form. FIGARO is designed to provide high sensitivity and good signal-to-noise performance, and is highly resistant to counter-measures. Since photons are used as the probing radiation, there is little to no residual activity in inspected items. When the accelerator is turned off, there is minimal residual radioactivity to impede handling and transport. The system hardware could be made sufficiently compact to put in a small truck or transport aircraft for rapid and flexible field deployment in response to changing threats, and a single accelerator could serve multiple interrogation portals at a given location.

PHYSICAL PRINCIPLES

The $^{19}F(p,\alpha\gamma)^{16}O$ reaction is exothermic with a Q-value of 8.115 MeV. The reaction proceeds via the population of excited levels of the compound nucleus ^{20}Ne which have large α-particle decay widths to excited states of ^{16}O. The transitions to the ground state and four excited states of ^{16}O take place through the emission of five alpha groups. The second, third, and fourth excited states of ^{16}O de-excite almost exclusively by the emission of gamma rays with energies of 6.129, 6.917, and 7.116 MeV, respectively. Thus the $^{19}F(p,\alpha\gamma)^{16}O$ reaction produces nearly mono-energetic, high-energy gamma rays of 6-7 MeV.

These gamma rays are sufficiently energetic to produce neutrons by photo-fission and/or photo-neutron reactions in fissionable material (e.g., uranium and plutonium), as the reaction thresholds are in the range 5.5-6 MeV. In addition, these gamma rays will

induce neutron emission from non-fissionable nuclear materials (e.g., deuterium, lithium-6, and beryllium). The (γ,n) thresholds are about 2.22, 5.73, and 1.85 MeV for these three nuclides, respectively. However, these 6-7 MeV gamma rays do not have sufficient energy to produce photoneutrons from common benign materials. The low resulting neutron background means that a DC accelerator can be used rather than a pulsed one, since we are not required to count delayed fission neutrons between pulses.

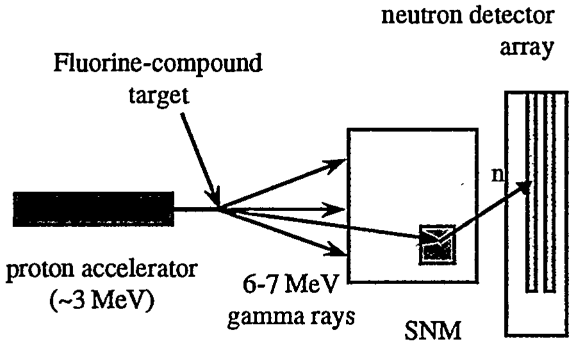

FIGURE 1. Schematic diagram of FIGARO operation.

Because the system would operate in a DC mode, and energy and time resolution are not required, one can use a very simple neutron detection system such as an array of moderated BF_3 or ^3He counters. We have chosen to use BF_3 because the energy released in the $^{10}B(n,\alpha)^7Li$ reaction is greater than that released in the $^3He(n,p)^3H$ reaction, allowing better rejection of low-energy gamma events and noise. Because very efficient use is made of both the accelerator-produced gamma rays and the neutrons generated by the interaction of those gammas, FIGARO has the potential to be a very efficient and effective detection system for all nuclear materials.

PROTON TARGET OPTIMIZATION

The requirements of a practical target for the production of energetic 6-7 MeV gamma rays from the $^{19}F(p,\alpha\gamma)^{16}O$ reaction are: (i) good mechanical strength and thermal conductivity to enable it to sustain significant beam currents at typical operating proton energies (up to 100 microamperes at 3 MeV, equivalent to 300 W of beam power) for an extended period of time with no significant deterioration; and (ii) minimal production of unwanted background neutrons from (p,n) reactions. For the latter consideration, reference is made to Table 1 (1). Although not shown in this table, the (p,n) threshold for mono-isotopic fluorine is 4.234 MeV.

Our earlier work in the development of this technology investigated the use of solid-compound fluorine targets (2). Both CaF_2 and MgF_2 in crystalline form were considered. These targets were found to be quite limited in their ability to survive even modest proton beam currents over reasonable time periods. Later a gas-target system employing SF_6 was tested and was found to be far superior to the solid targets for the present application. Initially, an existing target cell fabricated entirely from stainless steel was employed without modification. The gas-cell window selected was 5-micron-thick W foil. The tungsten windows thinned during use, most probably from being attacked by free fluorine radicals produced by dissociation of the target gas. This led to frequent catastrophic loss of the cell gas and failure of the accelerator vacuum system. Next, a 7-micron-thick Ni window was tested. While this arrangement was mechanically stable and contained the target gas reliably at pressures up to 35 psia and beam currents up to 2 μA, the yield of (p,n) neutrons was found to be unacceptably high above $E_p = 3$ MeV. Mono-isotopic Al has a relatively high (p,n) threshold (5.804 MeV) and also possesses desirable thermal and mechanical properties. Aluminum foil windows of both 15 and 20 micron thickness have been used with considerable success, with low neutron background and stable operation for extended periods at 13-15 psia and proton beam currents up to 4 μA. These are not fundamental physical limitations, so a target system capable of handling the more severe operating conditions expected of a fieldable system can and will be developed in the future.

TABLE 1. (p,n) Thresholds for Selected Elements*

Element	Isotope	Abundance (%)	p,n threshold (MeV)
Lithium	^7Li	92.41	1.880
Carbon	^{13}C	1.11	3.236
Oxygen	^{18}O	0.20	2.574
Sulfur	^{36}S	0.02	1.978
Chlorine	^{37}Cl	24.23	1.639
Chromium	^{53}Cr	9.501	1.406
	^{54}Cr	2.365	2.220
Manganese	^{55}Mn	100	1.032
Iron	^{57}Fe	2.119	1.647
	^{58}Fe	0.282	3.144
Cobalt	^{59}Co	100	1.887
Nickel	^{61}Ni	1.140	3.070
	^{64}Ni	0.926	2.496
Copper	^{65}Cu	30.83	2.167
Gold	^{197}Au	100	1.389

*Only isotopes with (p,n) thresholds below 3.5 MeV are listed

Because Al has a high (p,n) threshold, 0.1-mm-thick Al was used to line the existing gas cell walls, beam apertures, and collimators for reduction of neutron background. SRIM (3) calculations indicated that this thickness was sufficient to stop the most energetic protons used (about 4.25 MeV). The only region of the gas target assembly which was not protected from incident protons by an Al layer was the stainless steel beam stop at the end of the cell. However, SRIM calculations showed that the range of protons in SF_6 at 13 psia and 4.25-MeV proton energy is significantly smaller than the actual cell length. Therefore it is highly unlikely that any protons could actually reached the unprotected beam stop. SRIM calculations also indicated a small probability of scattered protons reaching the side wall of the cell, but it was (as mentioned above) lined with Al sufficiently thick to stop these scattered charged particles.

This approach reduced the (p,n) neutron background considerably although it could not be eliminated completely due to unavoidable residual neutron yield from (p,n) reactions at proton energies above their respective thresholds on the minor isotopes ^{13}C, ^{18}O, and ^{36}S found in the target environment. These neutrons appear to be produced primarily at the gas cell entrance. However, we found that the effect of these neutrons on the FIGARO detector system could be reduced by about a factor of 2-3 by placing hydrogeneous shielding in the vicinity of the gas cell entrance region in such a way that neutrons from the interrogated sample were not obstructed. Background reduction is important for improving system sensitivity.

NEUTRON PRODUCTION MEASUREMENTS

A benchmark setup was used to detect neutrons produced by (γ,n) and (γ,f) reactions using the gamma rays generated by the $^{19}F(p,\alpha\gamma)$ source. The measurements were performed using the 4.5-MV Tandem Van de Graaff accelerator at the Ohio University John E. Edwards Accelerator Laboratory. A new beamline section was fabricated to minimize the effects of (p,n) reactions from elements deposited along the beamline in other experiments. A gas cell containing SF_6 was mounted on the end of this beamline, with the neutron detection array centered on the entrance of the gas cell and about 16 inches away horizontally. Protons of energy 3.0 MeV were used with a 15-micron Al window, so that the proton energy entering the SF_6 was about 2.65 MeV. The neutron detector array consisted of 19 BF_3 tubes mounted in three rows in a polyethylene moderating assembly. The outputs from all the tubes in a given row were summed and recorded as a pulse height spectrum for later analysis. A simpler detection system suitable for field application could consist of just a lower-level discriminator and a scalar.

A large number of samples, both pure elements and compounds, were examined to determine whether γ-induced neutrons were produced. Figure 2 shows the neutron count rates for selected materials that were examined, including several nuclear materials and common benign materials. The results have been corrected for background and have been normalized for integrated beam current and target mass. The nuclear materials DU (approximately 4.1 kg of depleted uranium was used as a substitute for fissile materials), beryllium, and heavy water show a clear neutron signal. Lead, which might be used to shield gamma rays from the target, also produces neutrons as expected. No other common benign materials tested to date (a total of about 20) have shown any significant increase in neutron detection rate above background.

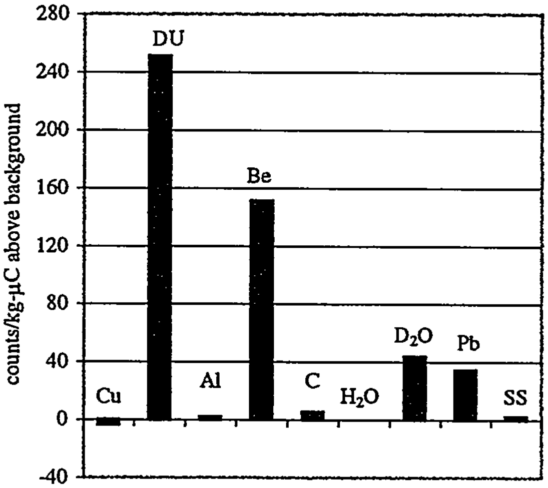

FIGURE 2. Normalized neutron count rates for selected nuclear and common materials.

In addition, studies have been performed to assess the resistance of the FIGARO technique to various countermeasures. Counter-measures to FIGARO could be medium- to high-Z gamma shielding to reduce γ-induced reactions, hydrogenous neutron shielding to prevent neutrons from reaching the detector, or some combination of the two. Both copper and lead gamma shields were investigated, along with neutron shields of polyethylene, borated polyethylene (BPE), lucite, and borax.

The results of some of these countermeasure studies are shown in Figure 3. Count rates of at least 10 counts/μC-kg above background are seen for even the case of one inch of lead in front of the DU with 3 inches of lucite behind. A more realistic geometry would move the gamma source, target material, and neutron detector further away from each other, but with increased detector coverage in solid angle. A reduction in count rate of no more than ten is expected. The detection of one kilogram of uranium with 1000 counts above background would require a proton beam charge of 1000 μC, which could be delivered by a 100 μA accelerator in ten seconds.

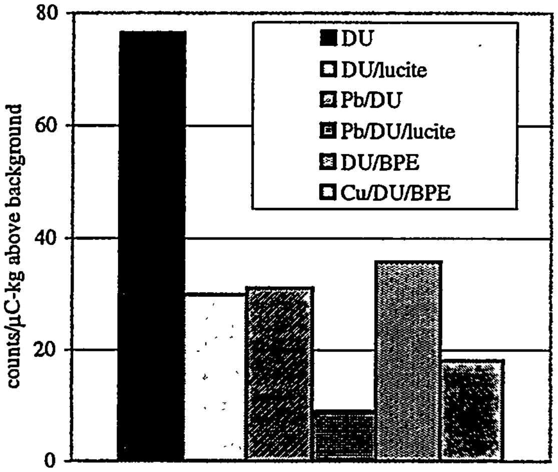

FIGURE 3. Normalized neutron count rate for trials employing selected countermeasures.

In a separate set of measurements, we examined the effect of placing the DU sample in an office wastebasket containing water. The DU sample was placed in the center of approximately 3-4 gallons of water. Results corrected for background and normalized to integrated beam current and target mass are given in Table 2. Even in this extreme case one can still see an increase in counts above background. It is important to arrange the test items so that the water minimally shields the detector array from the background neutrons produced near the gas cell entrance. If these are shielded, then the increase in neutrons counted due to the presence of the DU is offset by a reduction in the count of background neutrons. This points out another reason that the neutron background must be kept as low as possible.

Several measurements were also performed with the DU sample located inside a standard 9-inchx14-inchx22-inch carry-on suitcase filled with an assortment of clothes and personal articles. In each of several cases there was an excess of neutron counts above background sufficient to determine the presence of the sample (1000 counts above background) with an irradiation of approximately 50-250 μC.

TABLE 2. Results of Countermeasures Studies.

Case	Counts/μC	Counts/μC-kg above background
background avg.	35.3	-
DU	63.3	6.8
DU in water	44.7	2.3
water	36.5	-
background avg.	30.7	-
suitcase	15.3	-
suitcase + DU #1	118.4	21.3
suitcase + DU #2	97.1	4.3
suitcase + DU #3	122.6	22.3

CONCLUSIONS

Gas targets of SF_6 have proven to be strong sources of gamma rays from the $^{19}F(p,\alpha\gamma)^{16}O$ reaction. Aluminum windows have been used at E_p = 3 MeV currents up to 4 μA (12 W power) without failure. Benchmark measurements have shown neutron production from a range of nuclear materials with insignificant production from benign materials. The basic technique also appears to be resistant to countermeasures. Further directions involve the design of a high-power gas target and investigation of ways to use the technique to discriminate various types of nuclear materials.

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