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The detection of nitrogen using nuclear resonance absorption of mono-energetic gamma rays

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ABSTRACT

A new vacuum-insulated tandem accelerator capable of producing a 5-mA proton beam with energy up to 2 MeV was used to produce a mono-energetic beam of 9.17-MeV gamma rays from the resonant production reaction, $^{13}C(p,\gamma)^{14}N$, at 1.76 MeV. A graphite target enriched with ^{13}C capable of withstanding the proton beam power was designed and fabricated. The 9.17-MeV gamma rays were subsequently resonantly absorbed in ^{14}N via the inverse reaction, $^{14}N(\gamma,p)^{13}C$. The data acquisition system to measure the resonance absorption in nitrogen includes a BGO detector and a goniometer and collimator assembly that rotate around the axis produced by the intersection of the proton beam and the production target. The accuracy of rotation of the detector around the target is approximately 0.1°. The results of the resonance gamma ray absorption measurements are presented to demonstrate the feasibility of the method to sensitively and selectively detect high concentrations of nitrogen, comparable to those found in most explosives.

1. Introduction

The aim of the experiments is to construct a device that can reliably detect explosives. Since most explosives contain a higher concentration of nitrogen than common domestic materials, it is possible to use gamma ray resonance absorption in nitrogen to detect explosives. In this method the suspect object is scanned by 9.17-MeV gamma rays that are of the precise energy to be resonantly absorbed by the nucleus of nitrogen and dissipated by the other elements present. A comparison between the transmitted resonance and non-resonance spectra allows a determination of the presence of explosive material in the suspect object. [1–3]. The method is illustrated in Fig. 1.

In this case, a powerful electrostatic vacuum-insulated tandem accelerator (VITA) recently developed at the Budker Institute of Nuclear Physics has been used to generate monochromatic gamma ray interrogation beam [4]. Although originally developed for neutron-capture therapy of malignant tumors [5], the VITA is capable of generating a proton beam with a stable particle energy $\sim$2 MeV and a current of up to 5 mA in a continuous mode. An important property of the accelerator is the small energy spread of the proton beam ($<0.1\%$ $\Delta E/E$), as a result of electrostatic acceleration. Fortuitously, these were similar requirements to what was needed for a system based on nuclear resonance absorption in nitrogen.

A proton with energy 1.747 MeV incident on an enriched carbon-13 target is strongly captured in the resonant reaction $^{13}C(p,\gamma)^{14}N^*$ to form nitrogen-14 in the 9.172-MeV excited state. This composite nucleus recoils with a kinetic energy of 125 keV in the direction of the proton momentum. The lifetime of the level is short enough that the nitrogen nucleus de-excites in flight by the emission of a gamma ray whose energy depends on the angle between the direction of the emitted gamma ray and the momentum of the excited nucleus of the nitrogen. As a result of the Doppler effect, the 9.172-MeV gamma rays emitted at an angle of $80.7^\circ \pm 0.35^\circ$ have an energy of 9.172 MeV $\pm$ 122 eV, precisely what is required to be resonantly absorbed in nitrogen. A proton beam with high energy stability is required to produce the source of resonance gamma rays with high brightness, and this is precisely what the VITA provides.

Important scientific and technical problems were solved before the experiments could succeed, such as the production of a target enriched with ^{13}C, capable of sustaining powerful thermal and radiation loading from a proton beam, creation of a high-efficiency, high-resolution detector for 9.17-MeV gamma rays, with the appropriate data acquisition and analysis systems.

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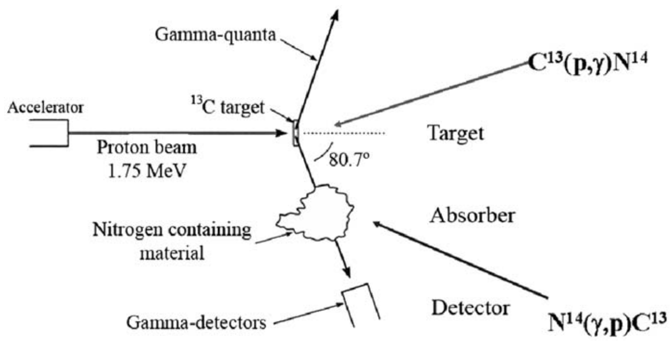

Fig. 1. The schematic of the experiment for detecting high nitrogen concentration.

2. Target design

The conditions under which the target must operate and survive are stringent, thus requiring minimization of the influence of several destructive factors: physical, chemical, and surface sputtering, and embrittlement.

Research of graphite stability under the influence of a proton beam was studied in detail in previous works, both experimental and theoretical (see for example [7]). The numerical calculations were carried out based on the Roth model [6], where physical and chemical are considered as coexisting simultaneously. This required that the region of optimum temperatures for a working target be in the range from 1100 to 2100 K for a proton beam energy up to 2 MeV.

Physical sputtering starts at proton energies higher than some minimum threshold, reaches a maximum, and then decreases, as a function of the increase of proton range in graphite. Chemical sputtering, on the other hand, depends mainly on target temperature. Its intensity reaches a maximum at target temperature $T\sim800$ K, and decreases with increasing temperature due to thermal decomposition of the formed hydrocarbons. Surface sputtering, in turn, is caused by the accumulation of gases in the surface layer and its diffusion accompanying graphite decondensation. At target temperatures greater than 1100 K, the total rate of target sputtering will not exceed 0.5 microns per day [7]. It should be noted that synthesis of methane occurs at a target temperature lower than 1100 K, and acetylenes synthesize at $T>1100$ K, the output of which grows a little with the increase of target temperature.

With the increase of graphite temperature, its evaporation increases as well, so the graphite vapor pressure reaches 1 atm at $T\sim3700$ K. At working temperature of the target up to 2100 K, vapor pressure is low enough to have a significant influence on vacuum conditions.

With calculations based on the above data, a thick graphite target enriched in carbon-13 was designed and fabricated with an equal concentration of ^{12}C and ^{13}C. Secondary electrons are suppressed with a high-voltage suppressor. Resonant gamma rays are emitted through a thin copper window. Proton current on target is measured. Heat allocated on the target surface is calculated based on the measurements of temperature and cooling water flow.

Two targets were used in experiments, the first of which was located too close (at 40 cm) to the accelerator and worked at a high power density ($\sim$3–5 kW/cm^2). Some scorches made by the proton beam on the target surface are shown in Fig. 2. Target temperature in the area of the beam was $\sim$3000 K according to estimations, so the rate of graphite evaporation exceeded 2.6 cm/s. This experience has been taken into account in designing the second target, which was located at a point in the beamline where an acceptable power density and surface temperature were achieved. The second target, shown in Fig. 3, has worked in a series of experiments totaling about 40 h at a proton beam current of $\sim$200 μA (up to 450 W/cm^2). Surface sputtering appears to be insignificant.

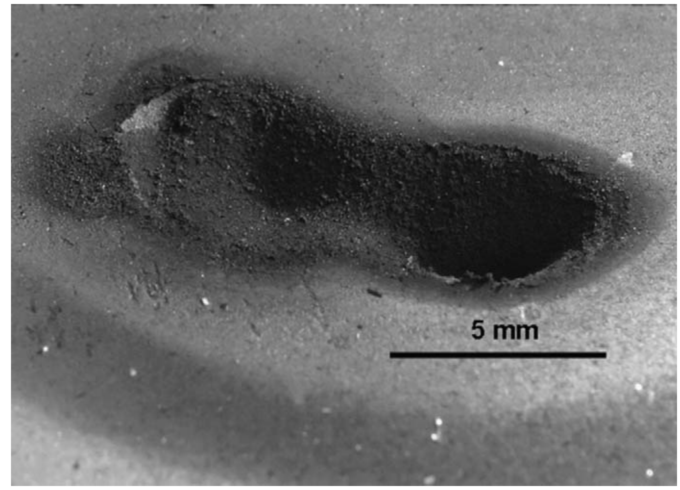

Fig. 2. The first target after two experiments, operating about 10 min in the 1.8-MeV proton beam in each experiment at 1 mA.

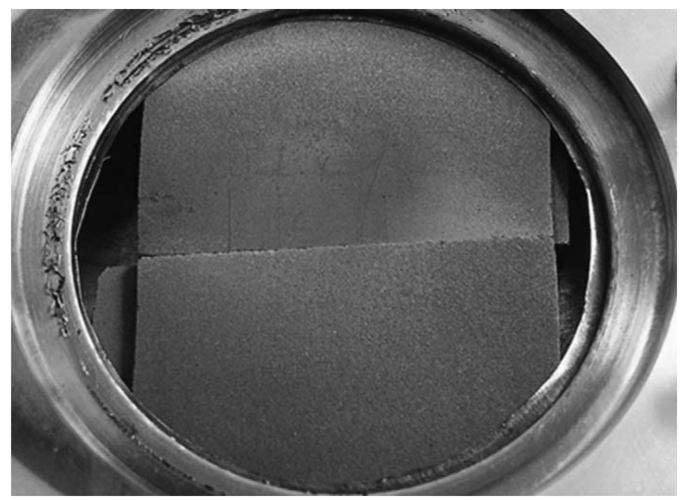

Fig. 3. The second target after $\sim$40 h in the $\sim$200 μA.

3. Gamma registration system

To perform the experiment similar to [8], a gamma ray spectrometer complex has been created (Fig. 4), including two scintillation detectors, positioned one over the other, inside a lead shield with $\sim$10-cm wall thickness as well as two lead collimators directed at the carbon target. A vessel containing liquid nitrogen is positioned such that gamma rays pass through the nitrogen and into one detector and miss the nitrogen in the other. To register the gamma rays attenuated by resonance absorption in nitrogen, the BGO scintillator detector is used; CsI is used as the other gamma ray detector intended to continuously monitor the intensity of the proton beam on the target. The entire system is placed on the goniometer, a mobile platform that rotates about an axis through the gamma ray-generating target. It provides for the measurement of both the resonant and non-resonant attenuation

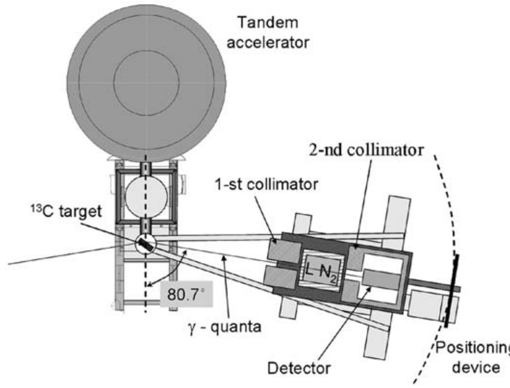

Fig. 4. Experimental layout.

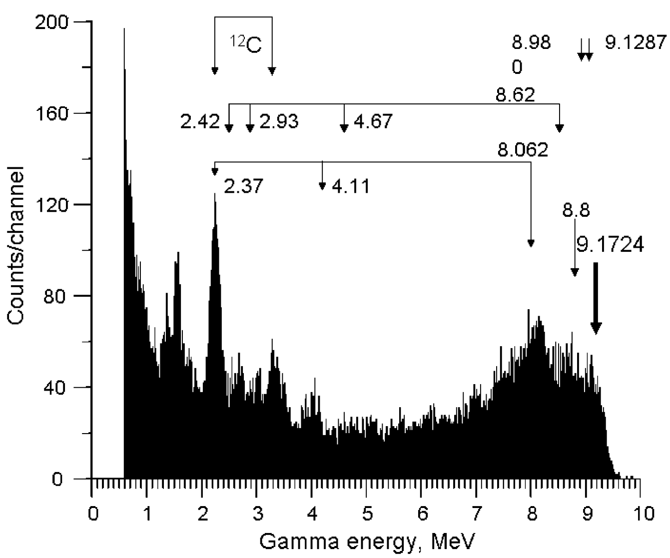

Fig. 5. Gamma ray pulse-height distribution measured from a thick carbon-composite target at 1800 keV proton energy.

in nitrogen by changing the angle. The accuracy of goniometer rotation is approximately ~0.1° and the total weight of the system is about 1 ton.

The gamma ray detectors use BGO ∅50 × 55 mm and CsI ∅80 × 80 mm scintillator crystals, each attached to Photonis XP3312B photo-multiplier tubes with the power supply optimized for spectrometry. All elements of each detector are covered with a metal shell and reliably shielded from the effects of magnetic fields and electromagnetic noise. Stability of the power supply voltage is approximately 0.1%.

The pulse-height analysis of the scintillation pulses from the gamma ray detectors is performed by two high-speed, computer-controlled, 4096-channel ADCs. The ADC resolution ranges from 50 to 4 mV. A special feature of these ADCs is zero dead-time and pulse per second. The software displays the spectrum as it accumulates in real time, saves and displays both saved spectra as well as set the acquisition time. When additional ADCs are installed in the same computer, it is possible to manage them synchronously and separately.

Preliminary calibration of the gamma ray spectrometer was carried out with the ^{60}Co gamma rays at 1173 and 1332 keV. The energy resolution of the BGO detector was determined to be 9.5% close in the region of ~1 MeV. Calibration precision was improved in the process according to identified gamma ray peaks in the actual acquired data.

Using computer modeling, it was shown that the BGO crystal had a full energy absorption for ~27% of the 9.17-MeV gamma rays. It was determined that an optimal crystal to use for the next series of experiments should be ∅80 × 100 mm in size. Such a detector would provide full energy absorption of ~70% of the 9.17-MeV gamma rays while providing the best possible energy resolution.

The temperature of the BGO crystal was observed to increase almost linearly by more than 1 °C during the experiment, resulting in reduced luminescence (1.34% per °C) and if not corrected will distort the energy pulse–height distribution. Since the width of the pulse–height distribution of the 9.17-MeV gamma rays is very narrow, even a small temperature drift in luminosity has a strong effect on the accuracy of the measurements. To compensate for the temperature drift, BGO temperature was monitored by a thermocouple and appropriate compensation in the analysis software was applied.

4. New method of normalization of the measured data and results of experiments

The spectrum of radiation generated by a thick ^{13}C target appears complicated (Fig. 5) in addition to the gamma rays generated by ^{12}C, which is also a major component of the target.

Gradually increasing the proton beam energy, the gamma rays associated with resonant production are encountered. Integrating the number of gamma rays in this 9.17-MeV region as the proton energy is increased through the resonant energy, an excitation curve for the given resonance is generated (Fig. 6). The threshold of the reaction is 1746.6 ± 0.9 keV [8], very narrow indeed. Thus, the slope of the excitation curve is defined entirely by the instability of the proton beam energy. As was established by the curve in Fig. 6, the proton energy resolution in the VITA beam is ~1% of the average beam energy.

In the process of carrying out this work, a new method for normalizing the measured data was suggested – normalization by the 2.36-MeV gamma ray connected with the reaction ^{12}C(p,γ)^{13}N. This line appears far enough from the 9.17-MeV line and, therefore, has a little susceptibility to changes in the resonant

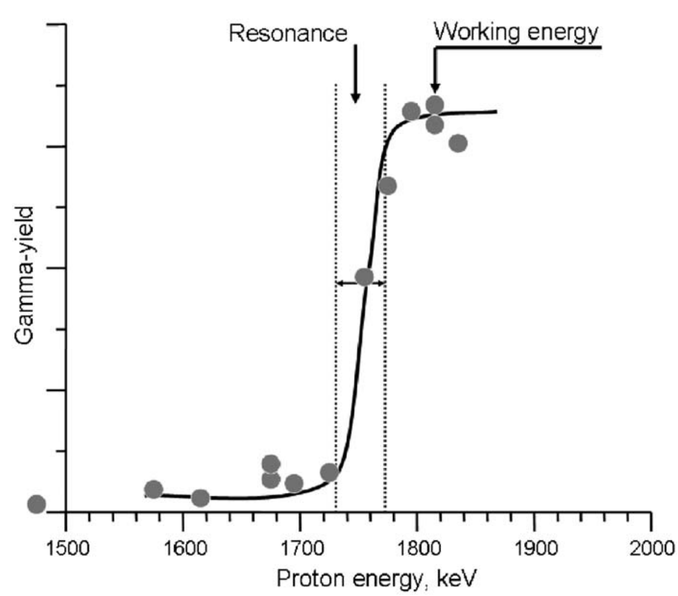

Fig. 6. Excitation curve for 9.17 MeV gamma rays from the reaction, ^{13}C(p,γ)^{14}N.

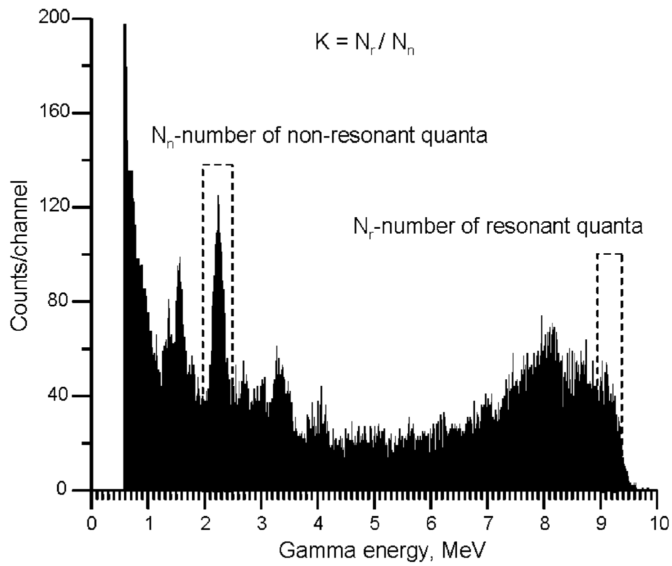

Fig. 7. Method of normalizing the resonance gamma ray intensity.

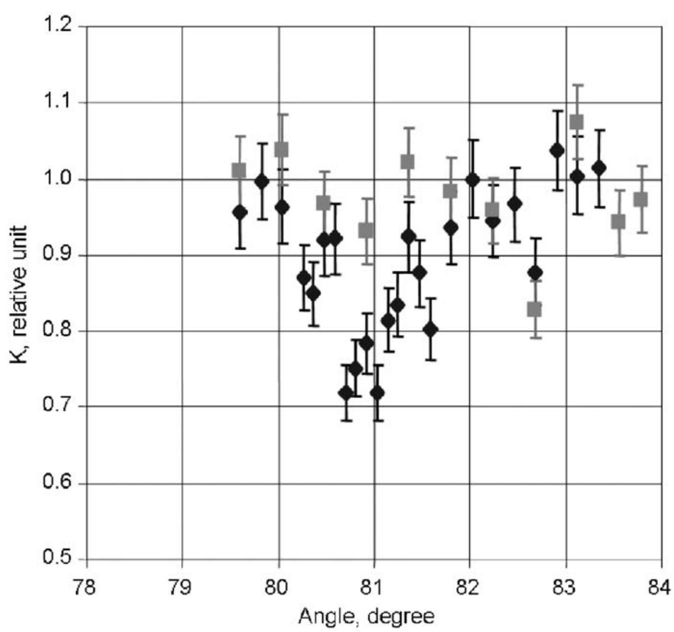

Fig. 8. Dependence of transmission coefficient K on the angle of the goniometer for liquid nitrogen-filled vessel (M) and water-filled vessel (O).

gamma ray output. At the same time, the output of 2.36-MeV gamma rays also depends directly on the intensity of the proton beam. Thus, this gamma ray can be used for normalizing the measured data because they are not resonantly absorbed in nitrogen. The transmission factor K, then, is the relation of the number of measured resonant gamma rays, N_r, to the number of measured non-resonant 2.36-MeV gamma rays, N_n (Fig. 7).

The pulse-height distributions of several gamma rays were measured and the transmission factor K was defined for different angular positions of the goniometer. A 38-cm-long vessel filled with either liquid nitrogen or water was placed in the path of the collimated beam of the gamma rays registered by the detector. For these two cases the dependence of the transmission factors K on goniometer position is depicted in Fig. 8. Noticeable attenuation of the resonant gamma rays is observed at an angle around 80.7° when the vessel is filled with nitrogen.

As observed in earlier experiments, resonance attenuation of 9.17-MeV gamma rays is still detectable when the vessel length is scaled down to 7.5 cm.

Thus, the results of the measurements presented here not only confirm the conclusions of previous research studies [9,10] but also show ways to improve the technique. First, it is shown that using a thick gamma ray-generating target simplifies its design and should increase the target lifetime. A more complicated distribution of gamma radiation in comparison with a thin target [8] is not an absolute obstacle. Second, a new normalization method is suggested and tested experimentally. This method allows us to discard the second gamma detector that naturally reduces the measurement time.

5. Conclusion

At Budker Institute of Nuclear Physics, the 2 MeV proton tandem accelerator VITA with vacuum insulation has been constructed and commissioned. It was originally developed to generate epithermal neutrons by means of the reaction $^{7}Li(p,n)^{7}Be$ at threshold for Boron Neutron Capture Therapy (BNCT) of malignant tumors. In the experiments described here, the accelerator is used for explosive-detection experiments by measuring the monochromatic gamma rays produced in the $^{13}C(p,\gamma)^{14}N$ reaction and resonantly absorbed in nitrogen.

A thick target is developed for generating 9.17 MeV gamma rays. It is made from the graphite enriched in ^{13}C and capable of withstanding a proton beam with high power density.

The diagnostic complex for detecting and analyzing the nitrogen-absorbing resonance gamma rays has been assembled and tested. It includes a goniometer with two collimators and lead-shielded gamma ray detectors.

A new normalization technique is presented and demonstrated. The dependence of the angular-dependent transmission factor on resonant gamma rays is determined for transmission through nitrogen and water. Noticeable resonance attenuation of resonant gamma rays is observed in nitrogen.

Thus, the first experiments conducted using a thick carbon target have demonstrated the generation of monochromatic 9.17-MeV rays and the reliable detection of their resonant attenuation in nitrogen.

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