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Calibration of a Direct Nuclear Activation Diagnostic Which Uses

Boron Nitride to Measure Energetic Deuterium Ions

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A direct nuclear activation diagnostic for the measurement of the kinetic energy of deuterium ions in the energy range of 1 to 3 MeV has been successfully developed. This diagnostic is based on the fact that the ratio of two different thick target yields which are generated by the same incident ion species can be a sensitive function of incident ion energy. Targets for this diagnostic are made of α -boron nitride (BN) and use the two reactions: $^{10}\text{B}(\text{d},\text{n})^{11}\text{C}(\beta^+)$ and $^{14}\text{N}(\text{d},\text{n})^{15}\text{O}(\beta^+)$. As long the BN material matrix remains constant, the ratio of thick target yields depends only on the deuterium ion energy and is independent of ion fluence and detector efficiency. Calibration of the diagnostic was accomplished by irradiating BN targets with deuterium beams of known ion kinetic energy and fluence. The calibrated diagnostic was then fielded to measure the voltage in a Plasma Opening Switch (POS). Electrical measurements of the POS voltage and the voltage inferred from the BN nuclear activation diagnostic were in good agreement.

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I. INTRODUCTION

Direct nuclear activation diagnostics are based on the phenomena that energetic ions can induce radioactivity within a target. By measuring the amount and kind of activity induced in a sample, information about the incident ions can be inferred. Nuclear activation diagnostics have been previously developed for measuring ion-beam fluences^{1,2}, voltages generated by pulsed power devices^{2,3}, and identification of impurities in beams or plasmas⁴. One previously developed technique to measure ion kinetic energy² is to measure the ratio of two different thick-target yields induced by a single ion species. This ratio can be a sensitive function of the incident ion energy over a limited energy range. In selected applications, the measured ion kinetic energy can be used to infer the operating voltage of a device. To make a useful measurement, however, the sensitive range of this diagnostic must correspond with the kinetic energy of the ions to be measured.

For this work, we were asked to measure the voltage in a Plasma Opening Switch (POS). Direct measurement of the voltage in the POS is difficult because the presence of energetic electrons, energetic ions, and x-rays all tend to adversely affect conventional voltage diagnostics. Nuclear activation diagnostics can still be applied in this situation, however, because they are insensitive to the presence of electrons and x-rays. For this application we selected the following reactions: $^{10}\text{B}(\text{d},\text{n})^{11}\text{C}(\beta^+)$ and $^{14}\text{N}(\text{d},\text{n})^{15}\text{O}(\beta^+)$. These reactions have several advantages. The most important advantage is that the ratio of the yields of these reactions is a

sensitive function of the incident ion energy over the anticipated voltage range of 1 to 3 MV. Another advantage is that there exists a readily available target material: hexagonal boron nitride (α -BN).

The half-lives of the product nuclides are also quite acceptable: 20.3 minutes and 2.0 minutes for ^{11}C and ^{15}O , respectively. The half-lives are long enough that the samples can be retrieved in less than one half-life, but short enough that counting times are not excessively long.

A complicating factor for this technique is the potentially interfering reaction: $^{10}\text{B}(p,\gamma)^{11}\text{C}(\beta^+)$. If the deuterium abundance is known, the individual contributions to the yield from the proton and deuterium reactions can be calculated². The correction is a function of ion energy² and, as a result, an accurate solution requires an iterative analysis. A more straight forward approach, which we use here, is to actively deuterate the source such that the proton reaction becomes insignificant relative to the deuterium reaction. To limit the contribution of the proton reaction to less than 1% of the total induced activity, the deuterium abundance needs to be increased to 0.3% or more.

In this paper we will first describe the basic principles of this activation diagnostic. In Section III we will describe the calibration of this diagnostic and present the resulting calibration curve. Finally, we will discuss the results of fielding this diagnostic to measure the POS voltage on the MITE accelerator. The results obtained with the BN nuclear activation diagnostic will be compared

with those obtained with electrical diagnostics which, which in turn, were verified with an ion time-of-flight (TOF) spectrometer.

II. THEORY

In this work we make use of thick-target , nuclear activation yields to infer a voltage. A thick-target yield is defined for targets that are thick enough to stop all incident ions within the target. The thick-target yield, $Y(E_i)$, for a monoenergetic beam of ions having a kinetic energy per ion, E_i , is equal to the number of product nuclides generated divided by the number of incident ions:

$$Y(E_i) = N_p / \Phi_i \quad (1)$$

where N_p is the number of product nuclei generated and Φ_i is the incident ion fluence. A thick-target yield curve can be obtained by measuring the thick-target yield as a function of incident ion energy. Since the yield depends on both the incident ion fluence and the ion kinetic energy, the measurement of a single thick-target yield is insufficient to provide any information about the incident ions other than possibly establishing the presence of an ion species. If one has independent knowledge of either the fluence or voltage, the other parameter can be determined from the activation yield. One possible source of such information is a second activation reaction.

Consider the ratio of two reaction yields:

$$Y_A / Y_B = (N_A / \Phi_A) / (N_B / \Phi_B) \quad (2)$$

where N_A and N_B correspond to the total number of reactants produced in reactions A and B. The ion fluences driving reactions A

and B are Φ_A and Φ_B , respectively. Over a certain ion energy range this ratio is a sensitive function of the incident ion energy. In general, there could be two spatially-separated targets and two different incident ion species driving the reactions. If either were the case, then there is little to be gained in using a second reaction because one still needs to know the fluence driving each reaction. If a single target material can be used, however, and if the same ion species drives both reactions, then $\Phi_A = \Phi_B$ and the ratio can be simplified to:

$$R(E_i) = Y_A / Y_B = N_A / N_B \quad (3)$$

In this situation, the ratio of yields is independent of the fluence on target and the ratio of thick-target yields, $R(E_i)$, is indicative of the incident ion kinetic energy. The fluence can then be calculated from the yield of either reaction.

This work is based on the two reactions: $^{10}\text{B}(d,n)^{11}\text{C}(\beta^+)$ and $^{14}\text{N}(d,n)^{15}\text{O}(\beta^+)$. By choosing BN as the target material, the two target nuclides, B and N, will be mixed on an atomic scale. Also, deuterons drive both reactions. The requirements for Eq. 3 are therefore met and measuring the ^{15}O to ^{11}C ratio will yield the incident ion kinetic energy over the sensitive range of 1 to 3 MV. It should be noted that both products are pure positron emitters, so the yields must be determined by using the differences in their respective half-lives, which is readily accomplished using standard half-life stripping techniques^{5,6}.

III. CALIBRATION

Calibration of this diagnostic consists of measuring the thick-target yields of each reaction as a function of incident ion kinetic energy. A calibration curve can then be produced by plotting the ratio of these yields as a function of deuteron energy. The experimental thick-target yield can be determined using the following expression:

$$Y = (C-B) \lambda / [1 - \exp(-\lambda t_0)] [\exp(-\lambda t_1) - \exp(-\lambda t_2)] \epsilon I_d \quad (4)$$

where (C-B) is the counts detected minus background in the counting interval which begins at time t_1 and ends at time t_2 . Both t_1 and t_2 are referenced to the end of irradiation. The time of irradiation is t_0 ; the product nuclide decay constant is λ ; the deuteron current is I_d ; and the efficiency of detecting the radiation from the product nuclide is ϵ . Since both product nuclides in this work are pure positron emitters, the detection efficiency is the same for each. As a result, the ratio of the two yields is independent of the detection efficiency. To determine the magnitude of the fluence, however, the detection efficiency must be known.

The calibration experiment was conducted using the Tandem Van de Graaff accelerator at the P-9 Ion Beam Facility at Los Alamos National Laboratories. Samples of BN were irradiated with deuteron beams of known energy and current. The activities induced in these samples were measured with a γ - γ coincidence, NaI detector system. To obtain the desired thick-target yield curves the deuteron energy was varied incrementally from 1.1 to 3.0 MeV.

Because BN is an electrical insulator, the ion current on target could not be directly measured. Instead, the ion beam current was measured with a Faraday cup just prior to target irradiation. It was then assumed that the current remained constant during the irradiation times which varied from 10 to 50 seconds. Measurements indicated that the current was constant to within $\pm 10\%$. Average currents ranged from 1 to 50 na.

After irradiation, each activated sample was then counted in a γ - γ coincidence system. The time between the end of irradiation and the start of the count was recorded. The γ - γ coincidence system detectors consisted of opposing NaI scintillators which were each 15.25 cm in diameter and 7.6 cm thick. The gap between the opposing NaI crystals was approximately 3 cm. Standard coincidence counting electronics⁶ were used to count the positron annihilation gamma rays. Counts were recorded using multichannel scalar techniques with a dwell time of one minute.

The measured optimal efficiency of this system for detecting annihilation gamma rays is about 23%, which corresponds to measurements made by other investigators⁷. Because the ratio of the yields does not depend on the efficiency, the actual efficiency of the system is not a critical parameter. In this work, the efficiency was not recorded, but was generally lower than the optimum value.

Because ^{11}C and ^{15}O are both pure beta-plus emitters, the individual activities can only be determined on the basis of the respective half-lives. Each activated sample was usually counted for approximately 40 minutes and the individual activities were extracted

using standard half-life stripping techniques^{5,6}. The products of the potentially interfering reactions $^{12}\text{C}(p,\gamma)^{13}\text{N}(\beta+)$ and $^{16}\text{O}(d,n)^{17}\text{F}(\beta+)$ were searched for, but, if present, were always below detectable limits.

Figure 1 shows the experimentally determined ^{15}O to ^{11}C yield ratio as a function of incident deuterium energy. This thick-target yield curve is the calibration curve for our diagnostic. The uncertainty of the ratio is at most $\pm 10\%$. The fractional standard deviation of the ratio due to counting statistics is actually less than 1%. The higher uncertainty is the result of the fact that if one chooses two different, but still reasonable subsets of data with which to calculate the ratio, the resulting, calculated ratio can vary at times by as much as 7%. This higher value we feel is more indicative of the true uncertainty in the ratio. We also irradiated multiple BN samples with deuterium ions having the same energy and found that the calculated ratios for the different runs agreed to within 3%. This agreement is further evidence that the total uncertainty is within our quoted $\pm 10\%$.

To apply this technique, one would measure a yield ratio of an unknown plasma or ion beam and then use the calibration curve to determine the ion energy. The ratio is a sensitive function of the voltage for all deuteron energies between 1 and 3 MeV, although it is clearly more sensitive for energies in the range of 2 to 3 MeV. For example, for incident ion energies in the range of 1 to 2 MeV, the estimated fractional standard deviation of the ion energy would be

roughly twice the fractional standard deviation of the measured yield ratio. For the ion energy range of 2 to 3 MeV, the fractional standard deviations would be roughly equal. This diagnostic could be used to measure deuteron energies less than the 1.1 MeV reported here, but we could not calibrate to lower energies because the Van de Graaff accelerator could not deliver stable beams below 1.1 MeV. We limited the upper energy to 3 MeV because of the potentially interfering threshold reaction: $^{11}\text{B}(\text{p},\text{n})^{11}\text{C}$. This reaction has a threshold of 3.02 MeV and is prolific. Even a small percentage of protons above 3 MeV in an unknown beam or plasma would significantly alter the ^{11}C yield and render the results meaningless.

We have also compared our results with those found in the literature. Thick-target yields based in part on neutron production data from the literature have been calculated by Young⁶ for the reactions: $^{10}\text{B}(\text{d},\text{n})^{11}\text{C}(\beta^+)$ and $^{14}\text{N}(\text{d},\text{n})^{15}\text{O}(\beta^+)$. Wohlleben and Schuster⁸ measured the induced radioactivity to determine the total cross sections of these reactions. We have taken the cross sections presented by Young⁶ and Wohlleben and Schuster⁸ and calculated yield ratios and compared them with our measured ratio. The three ratio curves are shown in Figure 2. Our ratio curve and that of Wohlleben have the same general shape. The values also agree to within the error bars of the data, but our results are systematically lower by about 20%. The curve generated from Young's calculations is different in both shape and magnitude. We believe that this may be due to the fact that he calculated some of the yields based on neutron emission experiments rather than those based on directly measured activities.

IV. APPLICATION

The calibrated diagnostic was subsequently used to measure the ion kinetic energy produced in a Plasma Opening Switch (POS) experiment⁹ being conducted with the MITE accelerator at Sandia National Laboratories. The pulse shaping capabilities of the POS in pulsed-power applications were being investigated. The POS plasma was produced with flash boards coated with a mixture of deuterated (98 at%) polystyrene and a colloidal graphite suspension in toluene. Over 100 BN diagnostic measurements of the POS plasma were made over the course of the experiment. In general, the voltages inferred from the BN diagnostic were of the magnitude expected from the POS and agreed well with inductively corrected voltages⁹ calculated for the POS region.

For roughly the last 35 POS measurements on which the BN activation diagnostic was fielded, an ion time-of-flight (TOF) spectrometer was fielded in the POS region as well. The ion TOF, which has an uncertainty of less than $\pm 10\%$, was used to verify the voltage from the electrical diagnostics⁹. The peak voltage as measured with the electrical diagnostics is compared with the BN-determined voltages in Figure 3. Comparison of the two measurements indicates that the voltage inferred from the BN diagnostic was consistently 10%-20% lower than the voltage as measured by the electrical diagnostics. The most likely explanation for the discrepancy is that the plasma has a distribution of ion energies and the BN diagnostic measures, not the peak ion energy, but rather the mean ion energy weighted by the rising cross sections. Numerical calculations of thick-target yields produced with

simulated ion energy spectra confirms that the BN diagnostic indicates a voltage that is usually somewhere between the first moment of the energy spectra and the peak voltage. There is experimental evidence that also supports this interpretation. On most MITE shots, two BN samples were used. On several of these shots, one sample was bare and the other was covered with 12.5 μm of aluminum foil. In these cases, the foil-covered sample almost always indicated a higher initial ion energy when corrected for dE/dx losses than did the bare sample. The difference between the bare and foil-covered samples is probably due to the ranging-out of the low-energy tail of the energy spectrum, which shifts the average energy towards the peak energy. Other possible mechanisms that could result in an apparent lower voltage are blow-off and ablation. Losses of reactants due to shocking of the graphite-like platelets and blow-off of surface dust cannot be readily accounted for. Calculations conducted with thermodynamic data obtained from the literature and MITE plasma energy densities (approximately $4\text{J}/\text{cm}^2$) indicate that significant ablation was unlikely¹⁰.

We also searched for the presence of several possible positron emitters that could interfere with our results. Surface analysis of target samples indicate that small amounts of oxygen(10at%), carbon(9at%) and calcium(1at%) were present. The bulk composition was 99% BN with 1at% calcium. It should be noted that in our targets the atomic ratio of boron to nitrogen was very close to 1:1. The use of targets with different stoichiometry would result in ratios of thick-target yields that differ from the ones presented here.

Although it appears that sample impurities are largely on the surface, calculations were conducted assuming a worst-case scenario in which the entire activated region of the sample contained 10%at oxygen and 9at% carbon. Based on cross sections and yields from the literature^{6,8}, we calculated the activities that would be induced via the reactions: $^{12}\text{C}(\text{p},\gamma)^{13}\text{N}(\beta^+)$ and $^{16}\text{O}(\text{d},\text{n})^{17}\text{F}(\beta^+)$.

Only the ^{13}N yield was found to be of possible significance compared with the ^{15}O and ^{11}C yields. We looked for the presence of ^{13}N and ^{17}F in the actual MITE data and saw no statistically significant evidence of either nuclide. This indicates that carbon-contaminated layer was thin. The possible presence of other interference isotopes was investigated by calculating the decay constants of the short-lived and long-lived portions of the experimental data. Significant deviations from the predicted decay constants for ^{15}O and ^{11}C were not found. BN samples were also irradiated with plasmas that were not deuterated and no significant positron activity was measured. This is strong evidence that proton and carbon reactions were not important in these experiments.

V. CONCLUSIONS

We have experimentally calibrated a direct nuclear activation diagnostic for the measurement of energetic deuterium ions in the energy range of 1 to 3 MeV . The diagnostic was fielded on the MITE accelerator and results of the BN diagnostic agree well with other, independent diagnostics that were fielded on the experiments. Results from the diagnostic are independent of the ion fluence and detector efficiency and, thus, should be reliable and portable to other users.

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CAPTIONS

Figure 1. The ratio of ^{15}O to ^{11}C thick-target yields as a function of incident deuterium ion kinetic energy. The uncertainty of the data points is $\pm 10\%$.

Figure 2. A comparison of the values we obtained for the ratio of ^{15}O to ^{11}C thick-target yields as function of incident deuterium ion kinetic energy (\bullet) with the ratios we calculated from data taken by Wohllenben and Schuster⁸ (*) and from calculations of thick-target yields done by Young⁶(x).

Figure 3. The peak POS voltage on MITE as measured with electrical diagnostics and verified with a time-of-flight ion spectrometer versus the voltages inferred from the BN nuclear activation diagnostic.

Figure 1.

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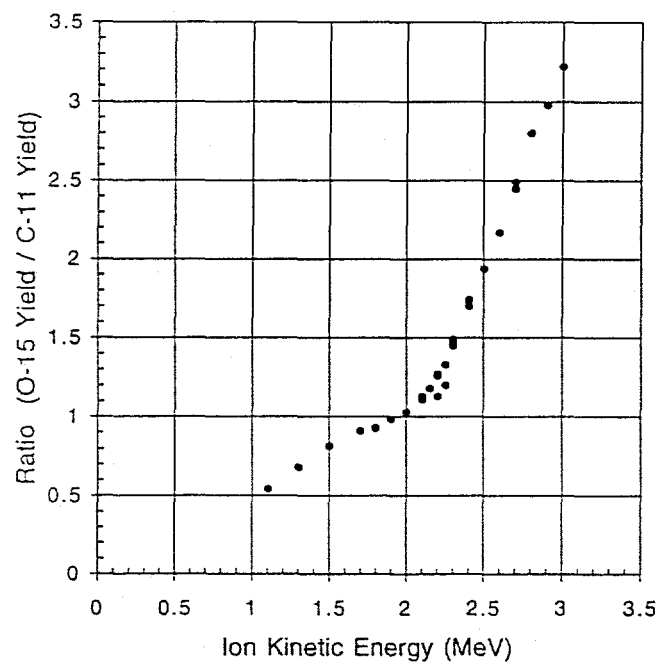


Figure 2.

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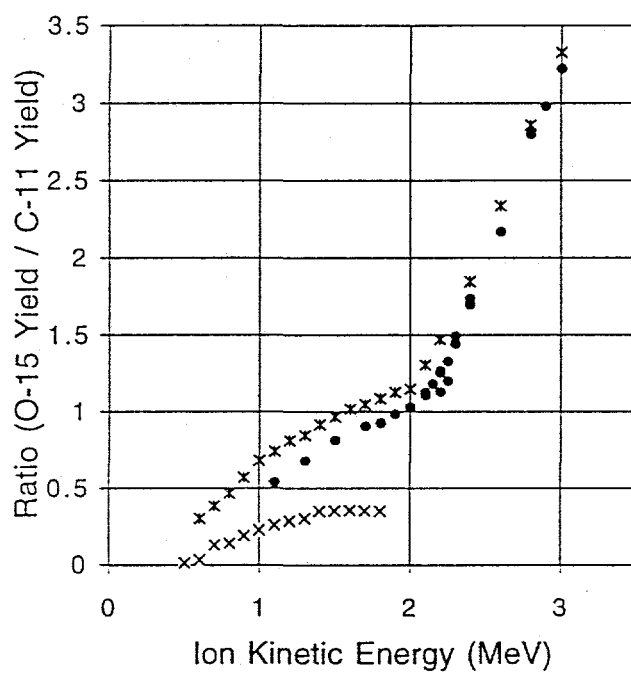


Figure 3.

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