Substance Detection using mono-energetic neutrons

August 4, 2004

Abstract

A method and apparatus for determining the nitrogen and oxygen composition of an arbitrary object in a non-evasive manner is presented. The object of interest is irradiated with fast mono-energetic neutron beams and the resulting pulse height spectra of scattered neutrons are measured using a SP2 spherical counter and unfolded to calculate the cross section of the object. The cross section is then compared to the well-known resonant cross section of nitrogen and oxygen. The potential of the method for making a bomb-detecting device is investigated.
I. Introduction

A non-evasive method that can determine the existence of Nitrogen and Oxygen in an arbitrary substance on an atomic scale, is of tremendous importance in biological sciences and Nuclear/Material engineering. The involvement of this method with fast mono-energetic neutrons is a promising idea since neutrons unlike charged particles are not influenced with long range forces such as the Coulomb force, thus they can penetrate a medium and propagate a long distance. Since different nuclei have different neutron scattering cross sections, different substances tend to scatter a fast neutron beam differently. This fact enables us to pose the problem at hand as an inverse-problem, i.e. by comparing the spectrum of the scattered neutrons of an unknown object to the spectrum of the scattered neutrons of a known object, we can make conclusion about the atomic composition of that unknown substance.

II. Theoretical Background and general setup

In this experiment, we use this method to implement a conventional bomb-detecting device. It is well-known that conventional bombs are made of substances that are rich in nitrogen and oxygen (see Fig. 1). It is also well known that the neutron scattering cross section of nitrogen has a narrow Lorentzian resonance peak at 433.6 kev while the neutron scattering cross section of oxygen is at 442 kev with a broader width (see Fig. 2).

The mono-energetic neutrons needed for this experiment will be produced by bombarding Beryllium and Lithium targets with mono-energetic protons created by RARAF Van de Graaf machine. The neutrons produced from the Be\(^9\)\((p,n)\)B\(^9\) and
Li\textsuperscript{7}(p,n)Be\textsuperscript{7} nuclear reaction will make up our neutron beam. It is important to note that these reactions will not produce neutrons that have mono-energetic angular distribution (cover a range from 0.35 Mev to 0.5 Mev), they create the 433.6 kev neutrons only at specific angles and these angles will be the places where the detection can be attempted. The thickness of our Lithium and Beryllium targets have been chosen to be about 100 nm, since a thicker target will create a neutron beam that is not mono-energetic. A target thinner than 100 nm is not desirable since it doesn’t create a neutron yield that is large enough for this application.
The detector is a SP2 proportional counter that is filled with hydrogen and has a pressure of 3 atm (absolute). It is positioned at an angle where the neutron beam has an energy that is equal to the energy at which the nitrogen and oxygen show a resonant cross-section. The detector can measure the energy and fluence (flux) of neutrons whose energy lies in 100 to 900 kev range.

The final setup is shown in fig. 3(below). The object to be probed is a bag which is moving in a conveyor belt.

III. The neutron detector

The detector is a SP2 proportional counter that is filled with high pressure hydrogen gas (3 atm absolute). The radius of this counter is about 1.9 mm. Its anode is connected to a high volt dc power supply and its cathode is grounded (see fig 4). The neutrons enter this high pressure region and collide with the hydrogen molecules in an elastic manner, the cross section of this collision is known to be isotropic relative to center of mass for neutrons that have energies in the region of our interest. According to Fermi(1935), “It is easily shown that an impact of a neutron
against a proton reduces, on the average, the neutron energy by a factor $1/e$”. The produced recoil protons have an energy distribution equal to,

$$P(E) \, dE := \int_{E}^{\infty} \frac{\sum(\xi) \cdot \phi(\xi)}{\xi} \, d\xi \cdot dE$$

(1)

where $P(E) \, dE$ the number protons with energy between $E$ and $E+dE$ in a differential volume of gas is given in the form of Volterra’s integral equation of the first kind where $\sum(E)$ is the cross section of hydrogen at energy $E$ and $\Phi(E)$ is the neutron fluence at energy $E$. These protons will travel in the gas under the influence of the electric field whose source is located at the anode wire. As they travel, they collided with other hydrogen molecules and cause more ionization in the gas. The ionized electrons will flow toward the wire, the rate at which the charge will increase is given by,

$$\frac{d}{dr}N(r) := -\alpha(r) \cdot N(r)$$

(2)

$$\alpha(r) := p \cdot A \cdot \exp\left(-\frac{p \cdot B}{N(r)}\right)$$

(3)

where $N(r)$ is the number of electrons ($r = $ distance from the wire), $p$ is the pressure of the gas, and $A$ & $B$ are Townsend phenomenological constant (for $H_2$, $A=0.0038 \, \text{mm}^{-1} \, \text{Pa}^{-1}$, $B=1.041 \, \text{mm}^{-1} \, \text{Pa}^{-1}$).

Eq. (1) represents the proton distribution in a hydrogen filled cavity that is infinitely long. Of course, since our hydrogen counter has a finite volume, so some proton recoils hit the wall before depositing their entire energy into the gas, and this brings in
the proportionality of the counter into question and equation 1 can not be used to calculate the neutron fluence. Snidow and Warren(1967) have accounted for this discrepancy and given the following equation,

\[
P(E) \, dE := V \left( F(E) \int_{E}^{\infty} \frac{\Sigma(\xi) \phi(\xi)}{\xi} d\xi + \int_{E}^{E_{\text{max}}} \left( \frac{d}{dE} \frac{R(E)}{\tau - E} \right) N(\tau(E) - R(\tau(E))) \left[ \int_{E}^{E_{\text{max}}} \frac{\Sigma(\xi) \phi(\xi)}{\xi} d\xi \right] d\tau \right) \, dE
\]

where \( v \) is the volume of the spherical counter, \( N(L) \) is the probability that the proton hits the wall after traveling a length \( L \), \( F(L) \) is the integral of \( N(L) \) and \( R(E) \) is the range that a proton travels in the gas before losing its energy completely. \( R(E) \) can be calculated by integrating Bethe-Bloch equation and is given by,

\[
\frac{dE}{dx} = -4\pi N_{A} r_{e}^{2} m_{e} c^{2} Z^{1} A^{1} \left( \ln \left( \frac{2m_{e} c^{2} \gamma^{2} \beta^{2}}{I} \right) - \beta^{2} - \frac{\delta}{2} \right),
\]

where \( N_{A} \) is Avogadros number, \( r_{e} \) is the electron radius, \( m_{e} \) is the electron mass, \( c \) is the speed of light, \( Z \) is the charge of our particle divided by the charge of electron, \( Z \) is the atomic number, \( A \) is the atomic weight, \( \beta \) and \( \gamma \) are the usual relativistic parameters, \( I \) is the ionization energy constant (~16Z^{0.9}) and \( \delta \) is the density effect.
Range vs. proton energy

Fig. 4

Range vs. alpha energy

Fig. 5
Fig. 4 and fig. 5 show the maximum range that a proton and an alpha particle can travel in hydrogen.

The charge collected at the anode is sent to a series of low-noise electronics that shape and record this electron pulse. Fig. 6 illustrates this process,

The pre-amplifier is a common base amplifier with a low input resistance and a high output resistance. The amplifier is a common-emitter amplifier whose output resistance is matched to the input resistance of multichannel analyzer. The amplifier integrates the pre-amp signal over a period of 6 micro-second and yields a Gaussian shaped pulse that has an energy equal to the energy of integrated signal, this Gaussian is then amplified ad sent to the multi channel analyzer where the peak value is detected and used to make a histogram which is called the pulse height spectra. This spectrum represents the proton energy distribution in the counter and will be unfolded using eq. 4 to find the neutron fluence spectrum. However, a calibration has to be done to determine the electronic gain of the counter, pre-amp., amplifier and multi-channel analyzer.
III. The calibration of the neutron detector

A small quantity of Americium-241 has been placed on the anode wire of the counter that decays spontaneously and creates a 5.6 Mev alpha particle. Since the decay rate is low (about 2 per second) and decay product is more or less mono-energetic, this source can be used to calibrate the counter and check its resolution. The resulting normalized spectra for different counter voltages is shown in figure 7,

![Normalized Spectra](image)

The peaks shown in figure 7 are due to the alpha particles which deposit about 1.17 Mev of their energy into the counter (see fig. 5) before hitting the wall. As we decrease the counter voltage, this peak will move to a smaller channel logically but as expected its general shape is preserved which confirms its proportionality. From this data, we can easily calculate $\alpha$ (the pre-amp gain) this enables to come up with an equation that describes the particle energy and channel number relationship. This relationship for the counter voltage of 1900 volts has been found to be,

$$E(\text{kev}) = \frac{44.384 \times \text{ch}}{\beta}$$

where $\beta$ is the amplifier gain as usual and ch is the channel number.
IV. Calibration of Van de Graaf using $\text{Li}^7(p,n)\text{Be}^7$ thresholds

A calibration of the proton energies produced by the Van de Graff is made. It is done by detecting the well-known thresholds of $\text{Li}^7(p,n)\text{Be}^7$ and $\text{Li}^7(p,n)\text{Be}^7^*$. According to Marion et al., the threshold proton energy for $\text{Li}^7(p,n)\text{Be}^7$ is $1.8811\pm0.0005$ Mev and the threshold proton energy for $\text{Li}^7(p,n)\text{Be}^7^*$ is $2.377\pm0.003$ Mev (see fig. 8). In order to carry out this calibration, a thin lithium target is bombarded by the proton beam in an ultra-high vacuum environment. A BF$_3$ neutron counter is placed at zero degree relative to the beam line. The number of neutrons produced by the lithium target (for a constant transferred proton charge) as a function of $\nu$ is shown in figure 9. $\nu$ is the NMR frequency of the Van de Graaf and is related to proton energy by a relationship given by $E_p = \kappa \nu^2$ where $\kappa$ is a constant to be determined. A line has been fitted into these points that has an intercept of 4661.0 khz which is interpreted as the threshold frequency. This frequency along with the well known 1.8811 Mev threshold energy, can be used to calculate $\kappa$. So, the relationship between NMR frequency and proton energy is given by,

$$E_p \ [\text{Mev}] = 8.66\times10^{-8}(\nu [\text{khz}])^2$$
V. Production of bomb detector neutrons using Li\textsuperscript{7}(p,n)Be\textsuperscript{7}

The Li\textsuperscript{7}(p,n)Be\textsuperscript{7} can be used to produce neutrons in energy range needed for the bomb detector. For a proton energy of 2.5 Mev, neutrons with energy of .434 Mev are produced at 90 degrees with a differential cross section of 15.1 MB/SR while for proton energy of 2.8 Mev, neutrons of .432 Mev are produced at 145 degrees with a differential cross section of 7.45 MB/SR (The cross sections are different by a factor of 2.017, see fig. 10 and 11 for further difference). The raw data and unfolded spectra of the 0.434 Mev and 0.432 Mev neutrons are shown in figure 12, 13, and 14 respectively. A peak with a large spread is seen in both cases since
Figure 12

Figure 13
Fig. 14

The difference between the Liskien and our data is summarized below. The results are very close each other which is satisfactory for calibration purposes.

<table>
<thead>
<tr>
<th>Value</th>
<th>Liskien &amp; Paulsen</th>
<th>My result</th>
</tr>
</thead>
<tbody>
<tr>
<td>En(145 deg.)[kev]</td>
<td>432</td>
<td>433.9 +/- 0.2</td>
</tr>
<tr>
<td>En(90 deg.)[kev]</td>
<td>434</td>
<td>433.9 +/- 0.45</td>
</tr>
<tr>
<td>The Ratio of differential cross sections at 145 and 90 degrees</td>
<td>2.027</td>
<td>2.13 +/- 0.15</td>
</tr>
</tbody>
</table>

Unfortunately, it is not possible to calculate the differential cross section from this data since the thickness of the lithium target is unknown but it will be calculated for the Be⁹(p,n)B⁹ reaction.
VI. Production of neutrons using Be\(^9\)(p,n)B\(^9\) At resonance

According to Gibbons and Macklin, the Be\(^9\)(p,n)B\(^9\) has a Lorentzian resonance at 2.563 Mev proton energy with a 100 kev spread.

This energy is of interest since the neutron yield of the reaction reached its maximum at this proton energy. The raw data obtained for proton energies from 2.3 to 2.5 Mev (steps of 0.05 Mev) and proton energies from 2.5 to 2.6 Mev(steps of 0.02 Mev) are shown below respectively,
Figure 14 shows the resonance seen at RARAF,

![Figure 14 Resonance of Be(p,n)B](image)

The peak is reached at 2.562 Mev which is in excellent agreement with Gibbons and Macklin data. The error bars are associated with the neutron counter resolution. This agreement is a further proof that our Lithium calibration is accurate to 1 kev.

Not much experimental results are available for the angular dependence of emitted neutron at resonance. It can be easily calculated using the basic laws of conservation of energy and momentum. This relationship is given by:

\[
E_n = E_p \frac{m_s m_n}{(m_n + m_r)^2} \left(2 \cos^2 \vartheta + \frac{m_r (m_r + m_s)}{m_s m_n} \left(\frac{Q}{E_p} + 1 - \frac{m_s}{m_r}\right)\right) + 2 \cos \vartheta \sqrt{\cos^2 \vartheta + \frac{m_r (m_r + m_s)}{m_s m_n} \left(\frac{Q}{E_p} + 1 - \frac{m_s}{m_r}\right)}
\]

Although this relationship is highly reliable, it is necessary for engineering purpose to be checked. At 10 different angles at resonance (\(E_p = 2.56\) Mev) the neutron energy has been measured. These angles are 0, 20, 40, 60, 76, 86, 100, 120, 140 and 160 degrees.
At 76 degrees, neutrons with 0.434 Mev are produced. The below plot shows the obtained pulse height spectra in an ascending order.
The deconvoluted spectrum in its ascending order is given below,
The measured and theoretical dependence of neutron energy on angle are shown below, as it can be seen there is a good agreement between the two values within the calculated error. (Note: the error in theoretical data are due to the fact that the counter subtends a 10 degrees angle. The error in the experimental data is mostly due to the large spread seen in some of the deconvoluted data).

![Neutron energy vs. angle](image)

**VII. The angular dependence of differential cross section of Be\(^9\)(p,n)B\(^9\) at the 2.563 Mev resonance**

Although there is experimental result for total cross section of Be\(^9\)(p,n)B\(^9\), no or very little experimental results are available on the angular dependence of differential cross section of this reaction, where the Be target is a thin target (~100 nm). One of the goals of this experiment is to measure this unknown angular dependence. The below plot gives the result of this calculation. The error bars are due to the fact that we don’t know the beam spot size (it is roughly approximated to have 0.3 +/- 0.1 mm diameter). It is assumed that the target has an infinitesimal thickness (in reality it is 100 nm thick).
Unfortunately, at 76 degrees where the bomb detector neutrons are generated, the cross section reaches its minimum.

**Acknowledgement**

The author wishes to thank Dr. Guy Garty, Dr. Gerhard Pearson, Mr. Steve Marino and Dr. David Brenner for their tremendous help. The author also wishes to thank RARAF and its staff for dedicating 80+ hours of their accelerator time to this project.
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