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Feasibility of the Use of Neutron Activation Analysis Techniques in an Underwater Environment

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FEASABILITY OF THE USE OF NEUTRON ACTIVATION ANALYSIS
TECHNIQUES IN AN UNDERWATER ENVIRONMENT

A Thesis
Presented to
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By
Michael Douglas Chick

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FEASABILITY OF THE USE OF NEUTRON ACTIVATION ANALYSIS TECHNIQUES IN AN UNDERWATER ENVIRONMENT

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FEASABILITY OF THE USE OF NEUTRON ACTIVATION ANALYSIS
TECHNIQUES IN AN UNDERWATER ENVIRONMENT

Michael Douglas Chick   December 2016   49 Pages

Directed By: Dr. Ivan Novikov, Michael Carini, and Phillip Womble

Department of Physics and Astronomy   Western Kentucky University

Elements when bombarded with neutrons emit a gamma ray that is characteristic of the

isotope that underwent a neutron induced nuclear reaction; this is known as neutron

activation. The characteristic gamma energy of an isotope can then be detected and

recorded. One can then analyze the gamma energies captured and determine the

elemental makeup of the sample. This form of analysis can be used in an underwater

environment making it potentially a valuable tool for agencies tasked with maritime

security of ports and waterways, or clean-up operations. This thesis will focus on the

feasibility of neutron interrogation using pulsed fast/thermal neutrons in an underwater

environment for detecting various chemical substances in metal containers. A

hermetically sealed, submersible container was used to test a d-T neutron generator's and

semiconductor detector's functionality underwater in regards to detecting such chemicals

as sulfur, nitrogen and chlorine rich materials.

Chapter 1 – Introduction

Section 1.1 The Criminal and Terrorist Threat

The potential for entities to do harm to the U.S. and the American way of life exists in the form of underwater smuggling and transportation of chemical, biological, radiological & nuclear (CBRN) threats and illegal contraband, but if we could detect these threats and contraband using applicable and practical technology we can continue to uphold our way of life. Illegal smuggling of such items is common place in American ports and off shore. There are multiple modes of trafficking used by smugglers to introduce weapons, narcotics, and other illegal cargo into the United States. In 2009, there were 24,737 kg of drugs seized that were transported via maritime means (Department of Justice, 2010). With most major maritime smuggling occurring through the Caribbean, see Figure 1.1.

Figure 1.1 Drug Flow into the United States (Figure courtesy of *Business Insider*)

One method traffickers' use is the "narco-submarine". The narco-submarine is a self-propelled, submersible vehicle which drug traffickers use to transport drugs into the United States, Mexico and Caribbean countries. Beginning in the 1990s, drug traffickers began using narco-subs for transporting their merchandise because of their difficulty to detect by means of radar, sonar, or infrared systems. On average one of these vessels can reach speeds of 15 miles per hours while transporting up to 10 metric tons (Ramirez, 2016). There is also an unmanned submersible, known as the narco-torpedo that also functions similarly to the narco-sub. It is a torpedo style container that is typically filled with narcotics towed behind another vessel. The narco-torpedo is released if authorities approach and is recovered at a later time (Ramirez, 2014). Though typically this medium for smuggling is used by drug traffickers, there are known ties between the drug trade and terrorism, thus opening up the potential for terroristic threats to be smuggled in this fashion as well. Admiral James Stavridis, former Joint Commander for all US forces in the Caribbean, Central and South America, wrote, "We need to be able to rapidly detect and interdict this new type of threat, both for its current effects via the drug trade, and – more troublingly – for its potential as a weapon in the hands of terrorists." (Department of Homeland Security, 2012).

Another commonly used method of underwater smuggling is the smuggling parasite container. These containers are welded, clamped or magnetically attached to the hull of a ship and contain drugs, weapons or other contraband. These parasite devices may or may not be known by the crew, they are attached to the vessel because they are hidden beneath water and those working with the drug organizations detach or open the boxes, and then move the cargo once the ship is in port (Anderson, 2011). Law

enforcement authorities, such as Immigrations and Customs Enforcement (ICE), use dive

teams to search the hulls of ships for such containers, as well as sweeps of the sea floor

and underwater structures for explosives and other suspicious activity (U.S. Immigrations

and Customs Enforcement, 2011).

Figure 1.2 Parasite Smuggling Box that was clamped to the hull of a vessel

(Figure courtesy of the *Washington Post)*

One further method of underwater smuggling used by traffickers today is using

shipping vessels with a false hull or false propeller shaft. Vessels with these have secret

compartments that traffickers use to store contraband in while the vessel seemingly

transports legal goods. A false hull and propeller shaft can be detected using ultrasound

scans (Hardesty, 2014). However, the ultrasound scan is unable to determine the contents

of such a compartment.

Divers are another threat as they can be used to smuggle contraband, or place

explosive devices or other dangerous items in and along the shoreline. Divers can

however be easily detected by sonar and are not likely to evade detection at any high value target, i.e. any major port in the United States. There is always the possibility these divers go undetected however, and as a result they may place some unknown device in an underwater setting that would need to be investigated.

Through each of these methods of smuggling and trafficking there is always the unknown aspect of what these containers may contain or these devices may actually be. This is an important aspect because inevitably human intervention will occur and before this intervention can occur, it must be known what the divers or other operators are coming into contact with. This is a major safety concern, as these containers while most likely contain illegal drugs have the possibility of containing chemical, biological, nuclear or radiological threats. These threats would have detrimental repercussions if improperly handled and dealt with.

Figure 1.3 ICE diver searching for smuggled threats on the hull of a ship (Figure courtesy of the *Washington Post)*

Section 1.2 Current Methods of Detection and Determent

The current system used by the US Coast Guard and other Maritime security forces to protect our ports from these smuggled threats is known as the Underwater Port Security System (UPSS). This two part system both detects and deters potential threats or smugglers by detecting, tracking, classifying and interdicting intruders, and inspecting hulls, piers, other underwater structures, and anything else that is underwater (Thomas, 2015). The first line of defense in the underwater port security system is the real time 3D sonar used to monitor the waters of a port even in zero visibility conditions. The second step in the UPSS is an Integrated Anti-Swimmer System. This system includes an underwater loud speaker designed to warn divers/swimmers that they are in a secure area, and it also includes an underwater shockwave system that can force divers to the surface.

Section 1.3 Principles of Neutron Activation Analysis

While the UPSS is effective at detecting smuggled materials and stopping divers from entering protected ports, when a possible threat is actually detected a team of divers enters the water to investigate the item. If that item is potentially harmful one must conduct a proper risk assessment regarding sending divers into the water to investigate. One possible technique for identifying the contents of a smuggled underwater container is by employing neutron interrogation, in the form of pulsed fast/thermal neutron analysis and prompt gamma neutron activation analysis, to identify the contents of said container. These techniques of nondestructive analysis are suited for this form of interrogation because of the quick detection results. Also the generally small size and ease of transportability of equipment used in this form of analysis lend it to be more practical in

field measurements where conventional methods are unlikely to prove practical (International Atomic Energy Agency, 2004).

The process of neutron activation analysis (NAA) involves a (n, γ) nuclear reaction or a neutron capture. "Activation Analysis" a term coined by Boyd (1949), involves neutron bombardment of a target element resulting in an inelastic collision with its nucleus creating a compound nucleus and immediate release of distinguishing gamma radiation. The nucleus becomes radioactive in this excited state. To de-excite, or decay, the nucleus emits distinguishing gamma radiation after the immediate or prompt gamma ray at a rate dependent upon the half-life of the of the now radioactive nucleus (Boyd, 1949). See Figure 1.4. This nuclear "fingerprint" can be used to determine the identity of the elements in the original nonradioactive material (Corliss, 1964).
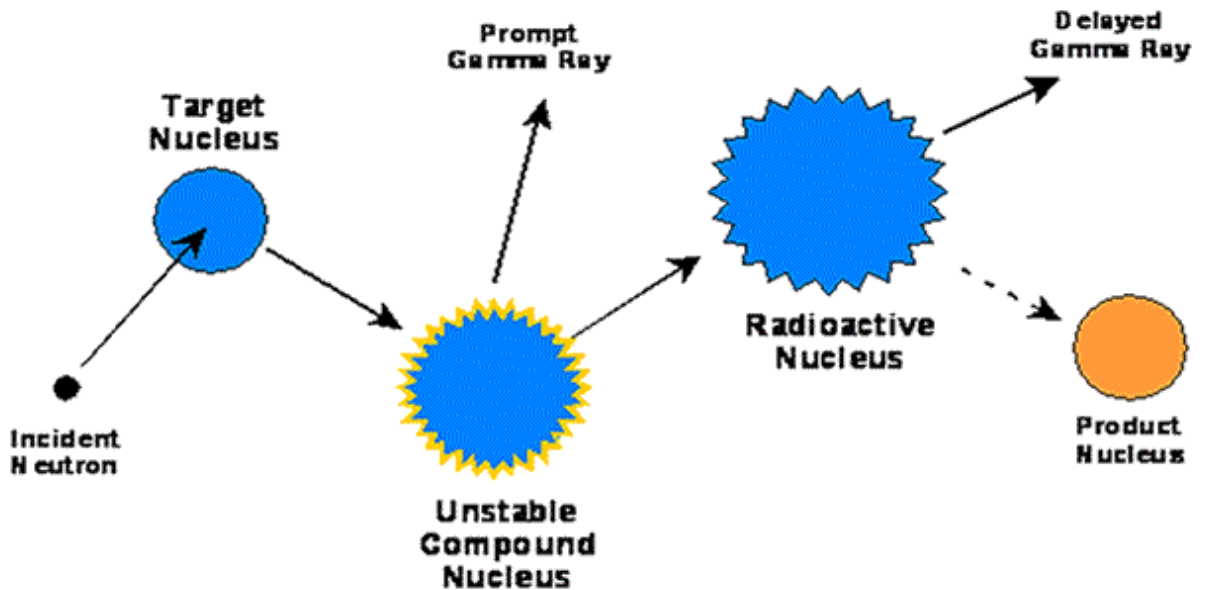

Figure 1.4 (n,γ) reaction

The radiation is distinguishing because each element produces gamma radiation at an energy level typically measured on the electron-Volt (eV) scale coupled with a half-life that is unique to each isotope. No other element decays at this energy level at this half-life making this detection method reliable and accurate in determining atomic make-up.

Neutron activation analysis is however, the overarching technique and this experiment focuses on variations within. One such variation is the technique of pulsed fast thermal neutron analysis (PFTNA). This technique employs the production of pulsed neutrons at 14 MeV which are needed for reliable measurement of elements such as C and O. These pulses occur for approximately 20µs at 14 MeV then tail off for approximately 80µs. This pulse occurs at a frequency of 10 kHz and then after a predetermined amount of pulses, there is a subsequent longer pause in production allowing for detection of gamma rays from activated elements. The 14 MeV neutrons, from the pulse, impinge on the sample object resulting in a number of nuclear reactions. The gamma rays from these reactions act as a fingerprint and can be used to identify the elements. The various reactions include inelastic scattering, (n,n'γ), (n,pγ), and neutron activation (Barzilov, Womble & Vourvopoulos, 2003). In between the pulses the remaining fast neutrons collide with the nuclei of light elements in the sample thus losing their energy. Once the neutrons have less than 1 eV they are captured resulting in the (n,γ) reaction mentioned above. The subsequent longer pause in the neutron production is implemented for the detection of elements that have been activated and require a halt in neutron bombardment to be viably detected.

The other variation of neutron activation analysis used in this experiment is prompt gamma neutron activation analysis (PGNAA), used in the detection of Cl. This

technique measures prompt gamma rays that are emitted from the excited state of the

sample nuclei within 10^{-14} seconds after neutron capture (Latif, Oura, Ebihara &

Nakahara, 2013). The prompt gamma emission is a result of thermal neutron capture

reaction that occurs during the moments in between the pulses of 14 MeV neutrons
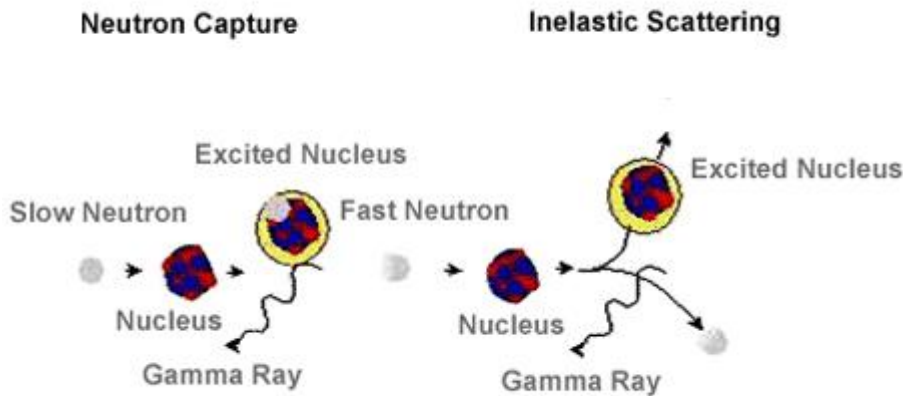
Figure 1.5 Nuclear reactions initiated by slow (thermal) and fast neutrons in PFTNA

during PFTNA. This occurs due to the slowing of the neutrons from collisions with

lighter elements within the sample. The neutrons are then captured resulting in a prompt

gamma ray which is then detected and used to identify and measure the element and

quantities based on the distinguishable prompt gamma energy.

Section 1.4 Neutron Production

To initiate the nuclear reaction, the source material must be bombarded with

neutrons. There are multiple methods for neutron production and different kinetic energy

levels at which the neutrons may be emitted. Neutron sources include reactors,

accelerators, portable generators, and radio-isotopic neutron emitters. They are emitted

as thermal (<0.5 eV), epithermal (>0.5 eV to <0.5 MeV), and/or fast (>0.5 MeV) neutrons.

As the neutron source is turned on, it begins to bombard the sample with neutrons causing the nuclear reaction. The sample is then exposed to the neutron bombardment for a period of time determined by the measurement sensitivity desired (Corliss, 1964). The longer the sample is exposed to the source, the greater the amount of the atoms will become radioactive, producing more gamma radiation making it more detectable. However, if the half-life of the sample material is short, it becomes unbeneficial to expose it to neutron bombardment for long periods of time because the radioactive isotopes will have decayed prior to being exposed to the detector.

Section 1.5 Gamma Ray Spectroscopy

Gamma ray spectroscopy, or gamma ray detection, is an analytical method that allows the identification and quantification of gamma emitting isotopes (Reguigui, 2006). This is how neutron activation analysis becomes a valuable tool for chemical analysis. Once bombarded by the neutrons, the sample emits the gamma rays that are detected and analyzed through this process.

> "The radioactivity produced by neutron reactions (most usually of the (n,γ) type) is analyzed by radiation measuring instruments. From such measurements, the radiation energy (or energies) and the half-life of the induced radionuclide can be determined with considerable accuracy," (Blanchard and Leddicote, 1959).

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Figure 1.6 High Purity Germanium Semiconductor detector with liquid Nitrogen (LN_2) cooling tank (Figure courtesy of Ortec)

The radiation measuring instrument, spoke of by Blanchard and Leddicote, used in this experiment is the semiconductor detector, see Figure 1.6. A semiconductor detector is used in gamma ray detection because as a gamma ray passes through the semiconductor it produces free electrons and holes, a lack of an electron where one could exist, the number of electron and hole pairs is directly correlated with the energy level of the gamma within the detector. The detection material of the semiconductor is arranged between two electrodes, and as the electron and hole pairs are produced they travel to the electrodes and produce a pulse that can be measured. The intensity of the resulting electrical pulses is a direct function of the intensity of the incident gamma ray. As the amount of energy required to create an electron and hole pair is known, and this energy is completely independent of the energy of the incident radiation, the number of electron and hole pairs can be used to determine the intensity of the incident radiation (Knoll, 2000). Knoll also states that in the semiconductor detectors, the variations in peak height are smaller and the resolution of the energy peak is greater, thus giving an advantage over other commonly used detectors such as scintillators.

Section 1.6 Quantifying the Data

Data collected in this manner is quantified by recording the counts, or number of

electrical pulses that occurred within the semiconductor material. One can cross

reference the counts across the different intensities with a table describing the gamma

energy levels of neutron bombardment induced radioactive isotopes, and determine the

atomic make-up of the sample. The electrical pulses are recorded as counts by digital

signal processing (DSP) through a multichannel analyzer. The digital signal processing

converts the electrical pulses to a digital signal and then assigns the signal to a

corresponding channel based on the energy level of the gamma ray and moment of

emission.

Based on the sensitivity of the detector it is becomes possible to determine

quantity of the element measured within the sample as it relates to gamma intensity. This

is of course dependent upon the efficiency and resolution of the gamma detector. The

data is quantified in graph form using counts as the (y) parameter and energy level (keV)

in the (x) parameter, see Figure 1.7. The photopeaks in the graph are a result of higher

counts at certain energy levels which are representative of the atomic make-up of the

sample. The height of the peaks is representative of the intensity of which the elements

produced gamma rays as a result of the neutron bombardment. The peaks are Gaussian in

nature and as such can be fitted to determine the statistical error in a given peak based on

a known gamma energy level. Error is dependent upon equipment used as different

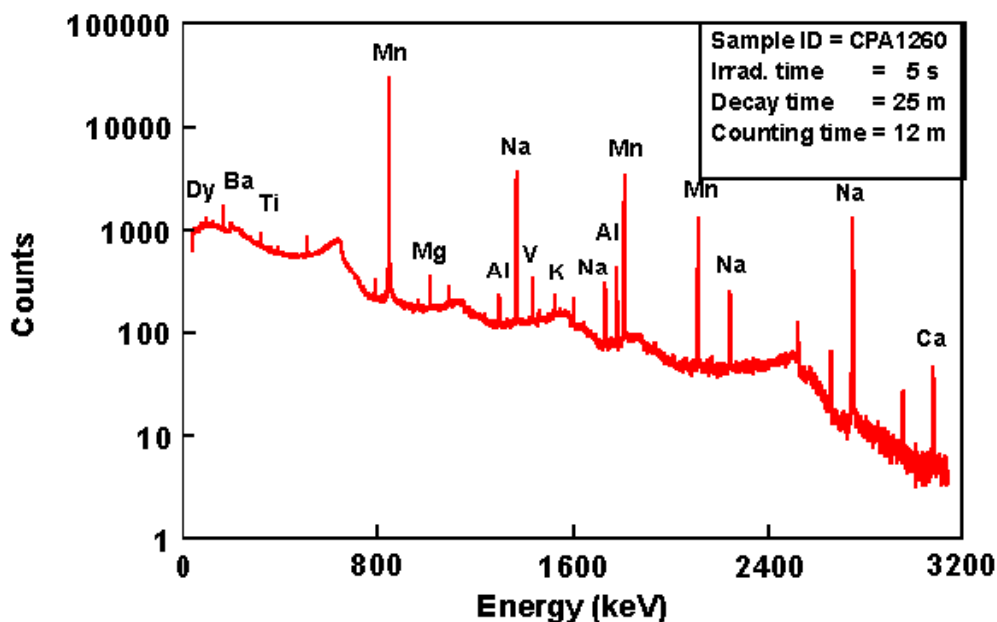detectors produce more or less defined photopeaks.

Figure 1.7 Gamma Ray Spectrum of Pottery (Figure courtesy University of Missouri Archaeometry Laboratory)

Section 1.7 Thesis Overview

The smuggling of narcotics is a constant issue faced by law enforcement agencies that are tasked to protect our borders. There are many methods and techniques criminals use to accomplish their goal of bringing these illegal items into the United States. Maritime smuggling of said narcotics has been an ongoing issue that has seen little to no decline since the war on drugs began. Due to the known connection between drug cartels and terrorist groups, it is possible that terrorists may attempt to use some of the techniques used by drug traffickers to smuggle weapons of mass destruction, which could be a chemical, biological, radiological or nuclear weapon, across the borders of the United States and our allies.

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Current detection methods for such smuggling are advancing, and many maritime smuggling operations are detected and stopped before they can reach the shore or port. However, human hands on interaction still plays a large part in these detection methods as an operator inevitably must handle any smuggling device located. This experiment is being conducted to determine the feasibility of neutron interrogation use in an underwater environment to determine the contents of said smuggling containers. In order to prevent the loss of human life due to contact with a possible threat located inside. If the contents of the container can be identified prior to human exposure, a proper risk assessment can be conducted and proper measures can be taken to ensure the safety of all involved in the detection process.

The goal of this experiment is to protect the welfare of operators of maritime security entities engaged in their lawful duties protecting American ports and waterways by providing them with a method of identifying unknown materials underwater. This is to be done by developing a practical method of employing neutron activation analysis based technique in an underwater environment. The remote operated vehicle would be used to place the equipment in range of the object to be analyzed, see Figure 1.8, for a period of time long enough to make a proper analysis.

Figure 1.8 Example of an underwater ROV (Figure courtesy of Underwater Engineering, Ltd.)

Chapter 2 – Experiment

Section 2.1 Overview

This experiment was conducted using a neutron generator in proximity to a target material in an underwater environment simulating field conditions. All analyzing equipment was contained within a sealed container and submerged within a pool, see Figure 2.1. The neutron generator was then energized and neutron bombardment of the sample would commence. The sample would then become activated by the neutrons and emit characteristic gamma radiation. The gamma rays were detected by a semiconductor detector, and data produced by the detector was analyzed via spectrum analysis software. That data was recorded as counts of gamma radiation, and graphed versus gamma energy level. This comparison produces photopeaks at a distinguishing gamma energy level of the detected sample material. The number of said counts would determine the effectiveness of the detection of the sample material within the underwater environment.
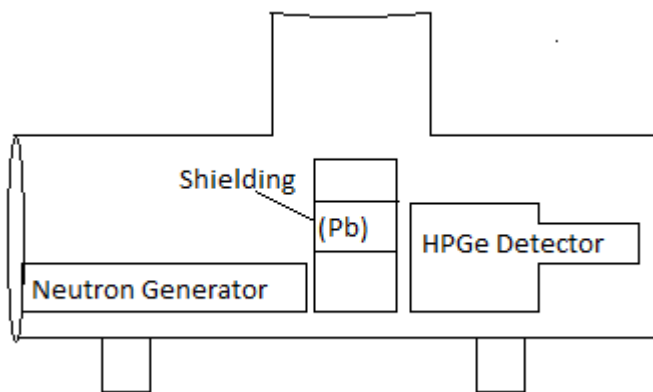

Figure 2.1 Representation of sealed container filled with the NAA equipment which was submerged underwater.

Section 2.2 Neutron Generator

There are multiple neutron sources that are used in scientific applications today, including nuclear reactors, radioisotopes, and accelerator based neutron reactors. Due to the practicality and portability required for this experiment, a sealed tube neutron generator was used. This compact device is a hermitically, sealed tube accelerator, see Figure 2.2. It operates by producing either a deuterium + deuterium (^{2}H + ^{2}H) or a deuterium + tritium (^{2}H + ^{3}H) reaction. This reaction occurs within the sealed, vacuum tube in which tritons (tritium nuclei) are accelerated into a deuterium-loaded target (Gow & Pollock 1960). The (^{2}H + ^{3}H) reactions result in the production of a helium isotope and a neutron. For this experiment 14 MeV were required; therefore, a neutron generator producing a deuterium + tritium reaction was used. The reaction is as follows: ^{2}H + ^{3}H → ^{4}He + n. This produces a ~14.1 MeV neutron (Chichester & Simpson, 2003). The requirement of fast neutrons is due to the fact that analysis of elements, such as nitrogen (N), oxygen (O) and carbon (C), are often difficult to determine using other analytical techniques employing thermal neutrons. The atomic ratios of C/O and N/O can be used to differentiate various materials (Papp & Csikai, 2011).

Figure 2.2 Sealed tube d-T neutron generator

The neutron generator produces the 14 MeV neutrons pulses isotropically, see Figure 2.3, and therefore all materials within range become activated that are not shielded. This produces background radiation that will be detected by the semiconductor. To mitigate the unintended neutron activation within the detector, lead (Pb) and high hydrogen content (H) shielding was placed separating the detector and the generator. Isotropic production is however beneficial because the neutron generator does not have to be aimed in any certain direction at the sample material, just within its proximity.
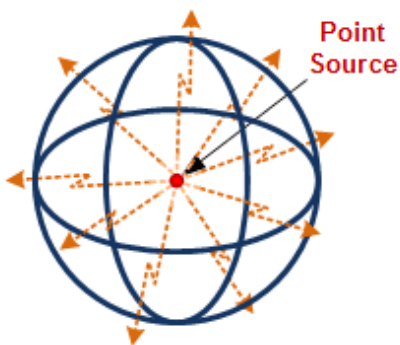

Figure 2.3 An example of Isotropic Production showing neutrons as they disperse in 4π steradians from the point source of the neutron generator

The maximum effective distance underwater between the neutron source and the

irradiated object is approximately 1 meter (Barzilov, Novikov & Womble, 2009).

Section 2.3 Semiconductor Detector

Many inorganic and organic materials have detection properties (Valkovic, 2015),

thus there are multiple options for which type of detector to use. Each type of detector

has its advantages and disadvantages. However in this experiment, a Canberra Cryo-

Pulse 5 High Purity Germanium (HPGe) semiconductor detector was used. The

advantage of this detector is its superior resolution compared to scintillation detectors

(Barzilov, Novikov & Womble, 2012). Superior resolution results in the well-defined

photopeaks produced when graphing the data relative to other scintillation materials, see

Figure 2.4. This type of detector has a relatively low band gap, see Figure 2.5, and has to

be cooled in order to reduce the electrical noise incurred as electrons cross the gap at

higher temperatures (Canberra, 2014). The cooling of the detector was achieved by a

built in cooling system utilizing a Canberra cryostat device. The system electrically cools

the high purity germanium to a temperature of approximately 77° K. This achieves the

high efficiency with the result being well-defined photopeaks in the 10 to 10,000 keV

range. During each analysis while the neutron generator was energized and bombarding

the target, the detector was also active, detecting the prompt gamma rays from the $(n,n'\gamma)$

reaction being expelled by the excited isotopes as neutrons impinged on the target

sample. This reading would be stored and then a second detection would take place,

utilizing the same detector, of the thermal neutron activation. This would be stored in a

second memory location. As stated above, the detector was shielded from the neutron

generator using lead shielding and high hydrogen content shielding. An HPGe crystal is

sensitive to the high energy neutrons, which cause detector damage (Tsoulfanidis &

Landsberger, 2010). A specific trial time was used to ensure enough counts were

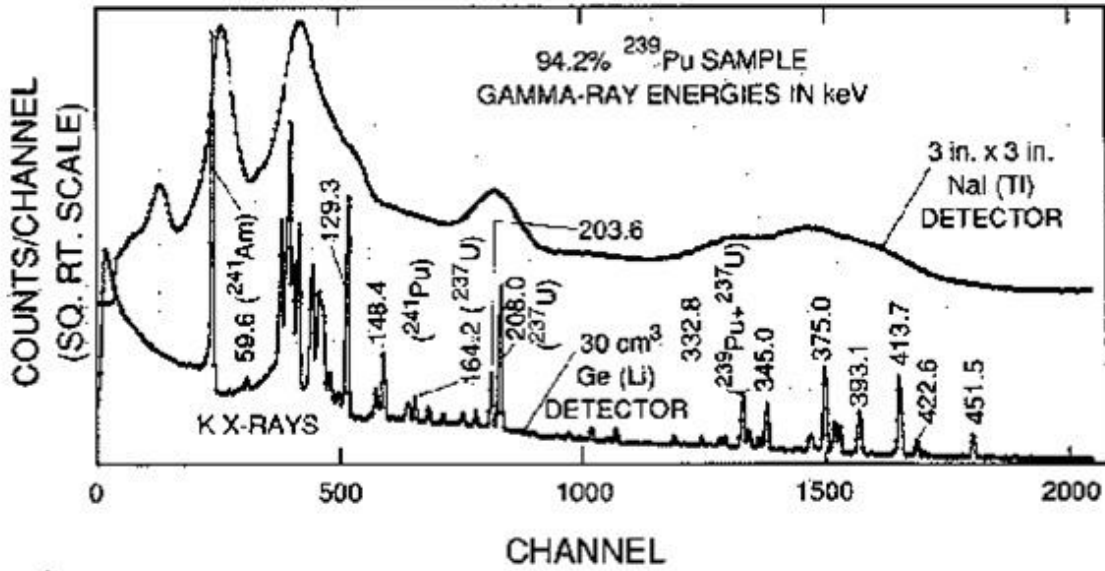captured by the detector to produce a quantitative analysis.

Figure 2.4 Comparison of photopeaks produced by a Germanium detector and a Sodium

Iodide scintillation detector (Figure courtesy of Texas A&M – Nuclear Security Science
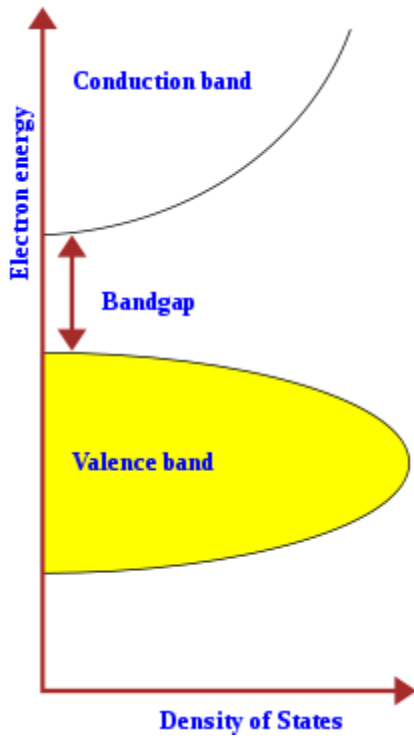
and Policy Institute)

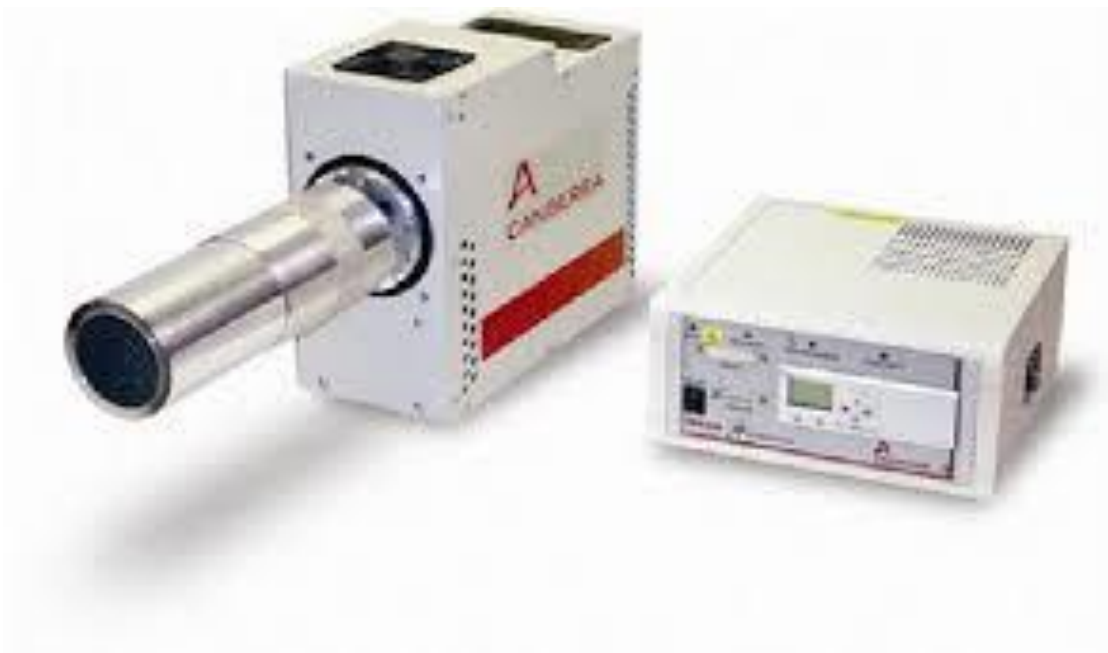**Figure 2.5** representation of a band gap in a semiconductor (no gap in a conductor)

Figure 2.6 Canberra HPGe Detector with electric cryostat

Section 2.4 Data Analysis

As the detector recorded energy counts of distinguishing gamma rays, it passed

this information along as a count per channel. Each analysis produced 8192 channels

with associated energy counts. The spectrum analysis software used to convert the data

from the detector to readable data was hardware and software developed for use with the

detector by Canberra in the Lynx Digital Signal Analyzer (DSA). The device records the

energy counts and assigns each set of counts a unique channel, and this is done in

multiple processes. The Lynx DSA is capable of recording dual sets of counts in its two

memory banks as it can read two sets of channels simultaneously. Initially, during the 14

MeV neutron pulse the system analyzes the signal from the detectors and registers the

gamma rays produced by the $(n,n'\gamma)$ reaction of C and O; it then stores that information

in particular memory bank. Subsequently during the time between pulses the Lynx DSA

analyzes the signal from the same detector and registers the gamma rays being emitted

from the reactions caused by the now thermal neutrons (the neutron capture reaction

(n,γ)), used to identify H, Cl, N, S, etc. This signal is then stored in another memory

location. The software within the Lynx DSA then converts signals into a counts given in

their respective channel. The counts are then in a readable text format.

The counts would then be formatted in a spreadsheet readily showing how many

counts per each channel as a counts versus channel table. It was then determined that the

distinguishing energy levels of hydrogen and oxygen, which had abundantly clear

photopeaks within the analysis due to the H_2O environment the experiments took place

in, would be used to calibrate the energy counts versus the energy levels, see Table 2.1.

The calibration was accomplished by determining which channel each of these

characteristic energy levels would be attributed to. This was apparent as the energy

photopeaks, and, therefore, count levels, for H and O were always the highest due to the

environment. Energy level was then graphed versus channel and fitted to a slope, using

mx+b = slope, see Figure 2.7. This slope was then used to graph energy (keV) versus

counts. The result being a graph showing photopeaks at characteristic gamma energy

levels for H_2O and the sample material being analyzed, see Figure 2.8.

Element	Energy (keV)
Hydrogen (H)	2223
Oxygen (O)	6130
O first release	5619
O second release	5108

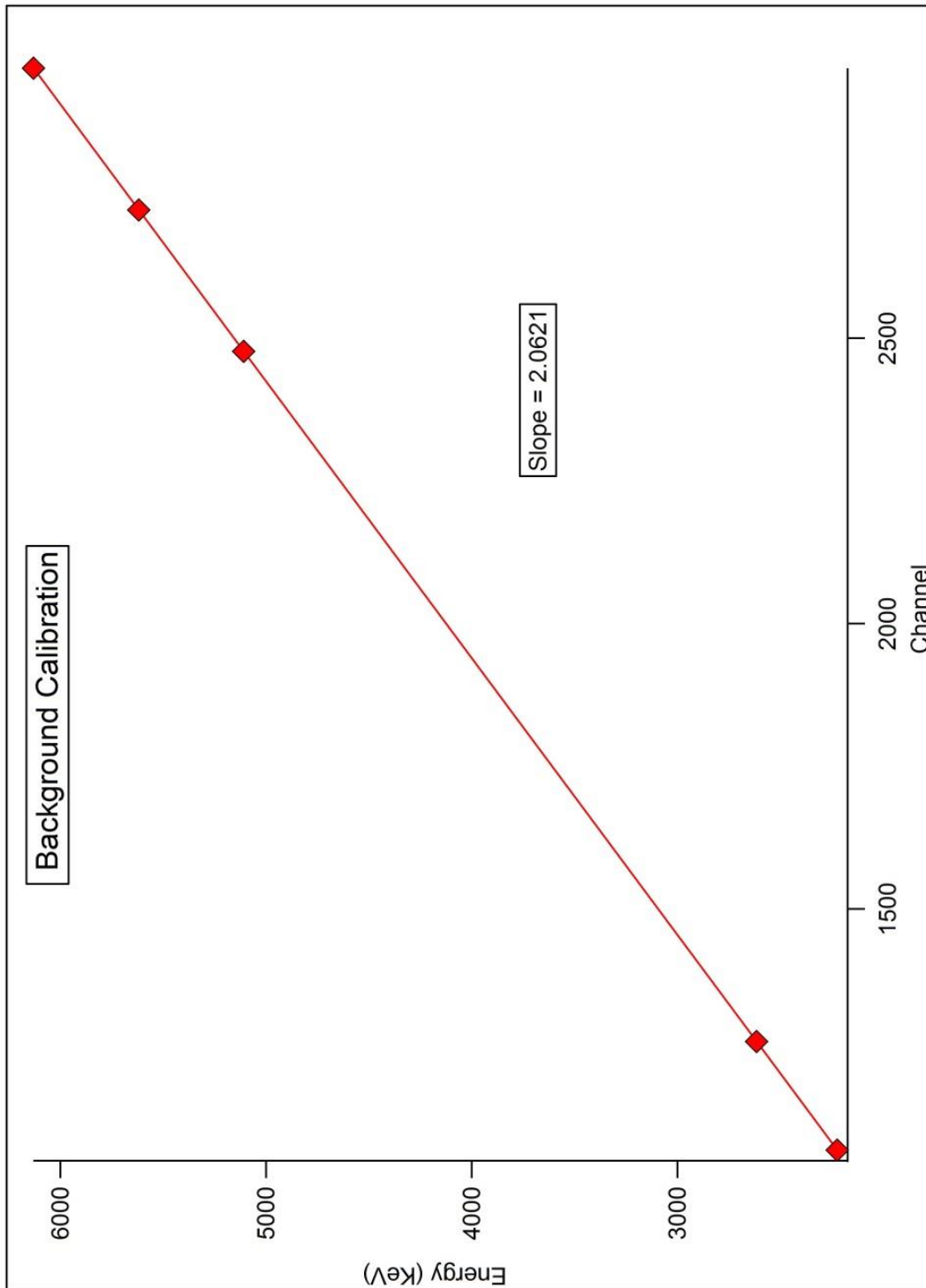**Table 2.1** Distinguishing Gamma Energies

Figure 2.7 Slope fitting calibration for characteristic energy levels vs counts
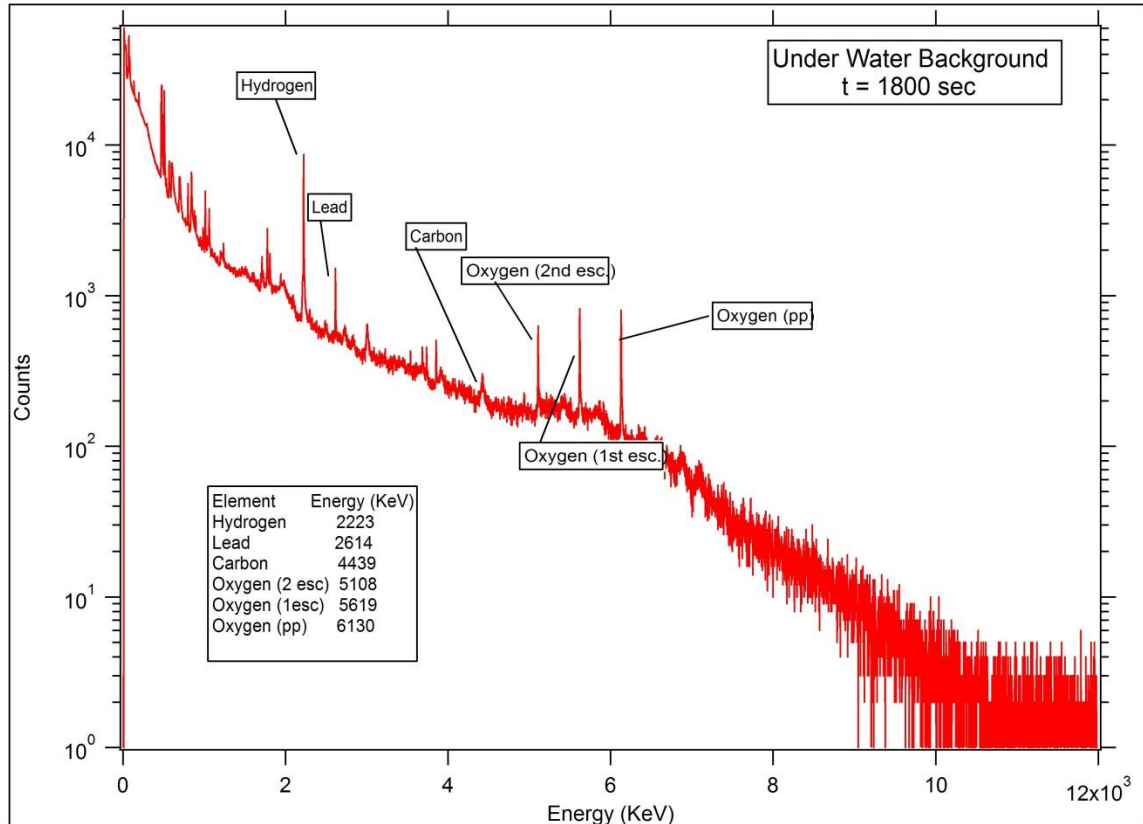
Figure 2.8 Example of Graphical Analysis of characteristic gamma energy

Section 2.5 Conducting the Experiment

This experiment was conducted in a rural location, partly a safety precaution and partly a convenience, where the simulated underwater environment was set up. This simulated environment was a standard diameter pool, approximately 7.5m and a depth or approximately 1.2m. The water contained within the pool did not contain any disinfectant or sterilizing chemicals to produce a more realistic field environment to include freshwater algae and bacteria.

The neutron generator and the HPGe detector were both placed within the sealed container and separated by lead and low Z shielding shielding. The lead shielding was in

the form of bricks and was approximately 10cm in width between the generator and detector. Digital control and power cords were run through an opening in the top of the container to the control module and power source. The container was then raised up and lowered into the pool using a mobile hoist system. The container was buoyant and had to be submerged using lead bricks that were fashioned to a tray at the bottom of the container. The container would then be positioned next to the target sample material.

The sample material was contained within a steel container that was also submerged using lead bricks placed atop it. The sample materials analyzed included NaCl, Sulphur (S), and Urea (N). Prior to analysis of these samples the detector would be calibrated using Co-60 and Cs-137. Chlorine is a powerful irritant that can inflict damage to the eyes, nose, throat and lungs, and at high concentrations and prolonged exposure it can cause death by asphyxiation (Romano, Lukey & Salem, 2007). Sulphur is another chemical surrogate representing sulfur mustard or mustard gas. It has the ability to form large blisters on any exposed skin or affect the respiratory and/or digestive systems (Center for Disease Control, 2013). Urea was used in this experiment to check system ability to detect presence of nitrogen in an underwater environment.

Each trial of the experiment was conducted for a predetermined time period. During each trial, the neutron generator would be energized for the duration and the detector would be detecting gamma radiation for the extent of the trial as well. The detector would then relay data to the Lynx DSA which would record said data. The data would then be analyzed and a determination of detectability for the sample material was made.

Chapter 3 – Results

Section 3.1 Calibration of the Detector

Typical gamma spectra produced in each trial of this experiment began with a calibration of the detector. Detector calibration was achieved through the use of radionuclides Cobalt-60 (Co-60) and Cesium-137 (Cs-137). The respective radionuclides produce gamma radiation at a known, constant energy, 662 keV for Cesium-137, and 1173 keV & 1333 keV for Cobalt-60 respectively. Calibration was achieved by observing the counts recorded by the detector at channels appropriate for the respective energy level. Through previous observation it was determined Cs-137 produced a defined photopeak with approximately twice the height of the two photopeaks produced by Co-60. Cs-137 resulted in approximately 1000 counts over a trial time of 1800 seconds, and Co-60 resulted in approximately 500 counts in both of its defined photopeaks at the same trial time, see Figure 3.1. Both radionuclides were measured simultaneously during detector calibration.

After measurements were taken, the data was placed in a table of counts vs detector channel. It was determined the channels with the highest number of counts were the respective energy levels of Cs-137 and Co-60. The energy level was then graphed against the channel number and fit with an $mx + b$ slope to determine the resulting energy of each detector channel, see Figure 3.2. The slope of 1.5986 was multiplied by channel numbers resulting in the basis for the energy levels on the *x-axis* of the graph seen in Figure 3.1.
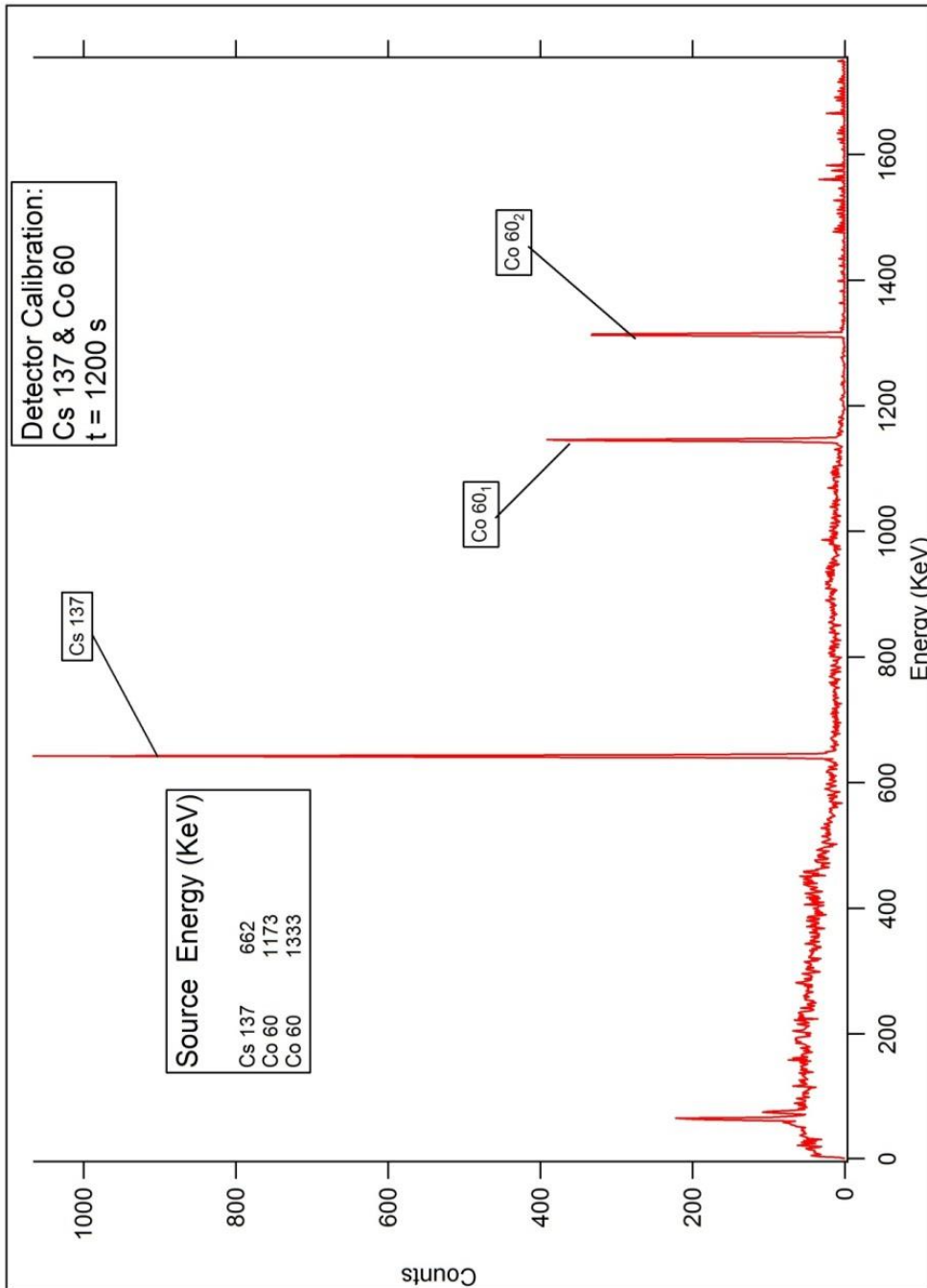
Figure 3.1 Detector calibration using Cs-137 & Co-60 showing their respective

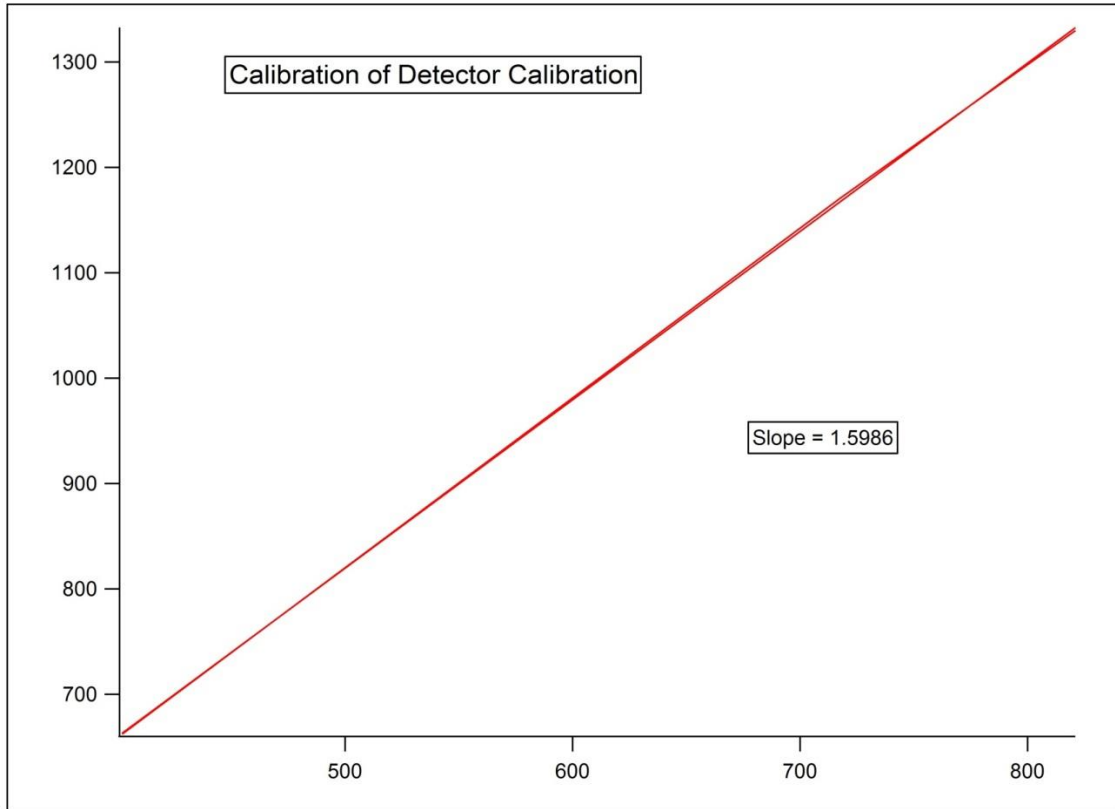photopeaks at 663 keV, 1173 keV, and 1333 keV

Figure 3.2 Calibration of gamma energy vs channel number

Section 3.2 Background Analysis

The first measurement taken after the detector had been calibrated was of the

inherent background gamma radiation, or noise, from the test environment. This

measurement was taken with the equipment submerged in the simulated underwater

environment. Background radiation had to be taken into consideration when determining

the results of the experiment due to this source of radiation being measurable which may

skew the data in some instances. For this trial there were no artificial sources of

radiation, beyond the neutron generator, applied by the conductors of the experiment nor

were there any sample materials applied to be measured. The resulting spectrum shows

Hydrogen (H), Oxygen (O), Lead (Pb) and Carbon (C) in high concentrations, see Figure

3.3. Hydrogen and Oxygen are in abundance due to the underwater environment in

which the experiment was conducted. Lead was measured as it was used for shielding

the detector from the neutron generator which would damage the detector and it was also

used to submerge the detection equipment. The presence of carbon was detected due to

its presence in the surrounding environment and shielding materials used.

As with all of the trials once the radiation counts were determined, the counts

must be calibrated against the gamma energy levels, see Figure 3.4. The slope of this line

= 2.0621 creating a proper fit for the counts vs energy graph. Figure 3.4 is an example of

the slope fitting used to fit the gamma energy counts to the corresponding energy. This

was part of each analysis. For this and all subsequent analyses hydrogen and oxygen

counts were used to determine the relation of detector channel to energy level. The

calibrations were conducted by determining the first extreme photopeak was hydrogen

and the subsequent three extreme peaks were the different releases of oxygen and fitting

the slope that way.

Once the calibration has taken place, the information can be analyzed. The area

of the photopeaks is equivalent to the gamma intensity being detected for said element.

When determining gamma intensity the following formula is used:

$$I_\gamma = \phi \times \sigma \times a$$

Where I_γ is the intensity of the incident gamma, ϕ is the incident neutron flux ($n/m^2/s$), σ

is the nuclear cross section of the element, and a is the atomic quantities of an element.

Given that the measured area of the photopeak is the gamma intensity, and assuming a constant ϕ, we can use the following equation to determine the atomic ratios.

$$a = I_\gamma/\sigma$$

In particular C/O and C/N are of interest. The atomic cross sections of each of the three elements are seen in Table 3.1, as well as the cross section of S and Cl which will also be discussed.

For these determinations gamma intensities are found by measuring the area of the photopeak using a peak area function in Igor Pro analysis software. The area of the peak is determined by fitting the data with a baseline to account for background noise, subtracting said baseline and measuring the area of the remaining peak at the gamma energy of interest. Table 3.2 represents the gross and net areas of peaks that will be used in further analysis in this experiment.

Element	E_n	Cross Section (mb)	γ-ray (keV)
C	14 MeV	184.7	4438
N	<1 eV	75.45	5110, 10833
O	14 MeV	82.54	5108, 5619, 6130
S	<1 eV	523.53	5420
Cl	<1 eV	33070.23	1165, 6110, 6619

Table 3.1 nuclear cross sections
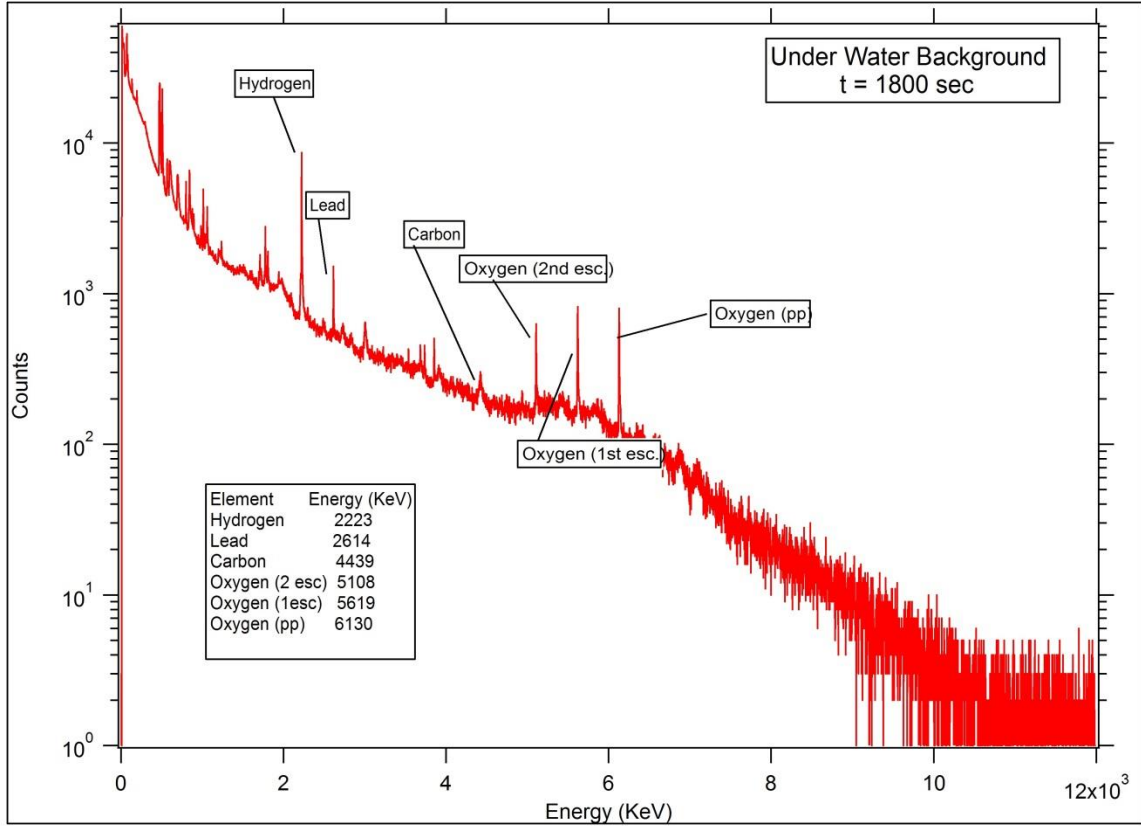
Figure 3.3 Graph of the background radiation with no sample material present

Background		γ - intensity	
Element	keV	Gross	Net
C	4432	1348	185.33
N	10833	----	----
O	6130	3840	2347.1
S	5420	----	----
Cl	1165, 6619	----	----

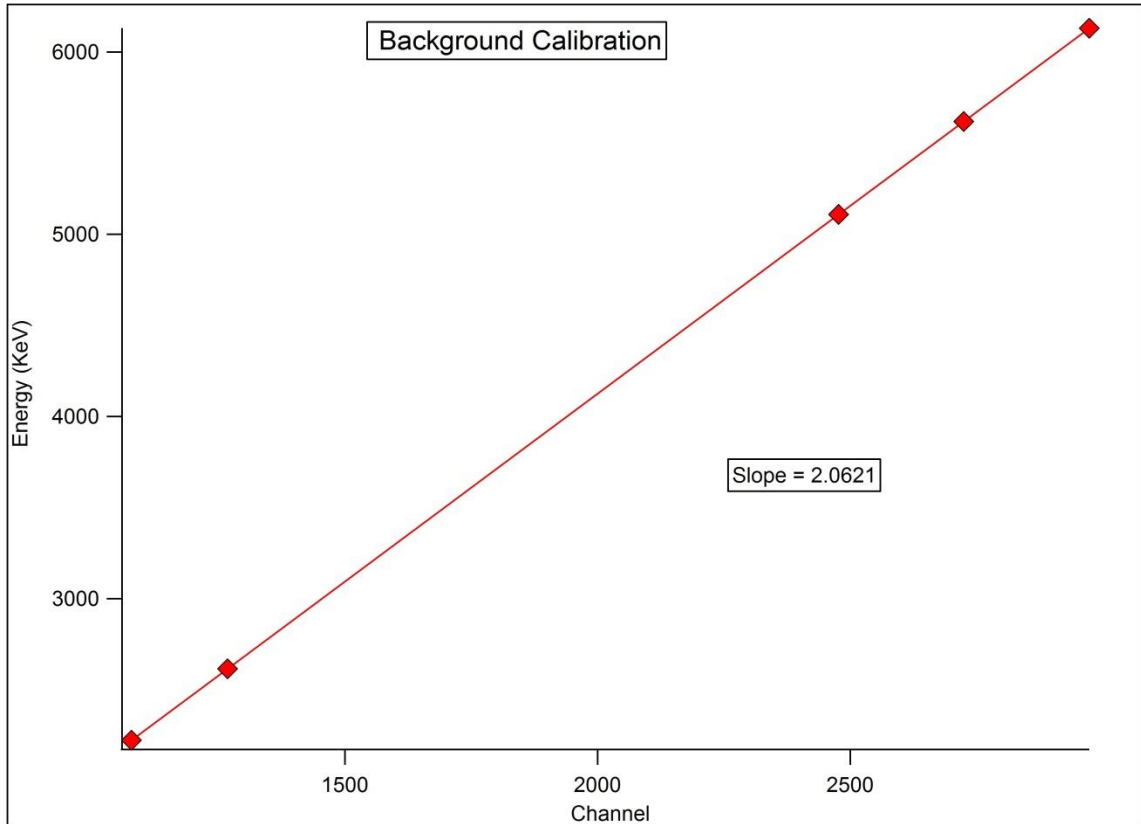**Table 3.2** Peak areas of observed elements in the background

Figure 3.4 Energy to channel calibration for the background radiation or "noise"

To determine whether an element was present in the background or not, a minimum detection limit was established. This limit was based upon the background noise of the experiment. As mentioned above, a baseline measurement was formed to account for this background. The minimum detection limit (σ) for this experiment is equal to the square root of the baseline using the full width of the photopeak.

$$\sigma = \sqrt{I_B}$$

Where $I = \gamma$ intensity and $B =$ baseline in this equation. The photopeak is identified and its area measured. Then using the width of the peak, the area of the baseline is determined. If the net area of the photopeak produced by an isotope is larger than σ it is

deemed as a detectable result. All figures listed in Table 3.3 were detectable using this method of determining minimum detection limit, and this is true for all other photopeak detection in this experiment.

Another factor to consider about detectability is the uncertainty of the measurements taken by the detector. While the detector is calibrated using constants, there is still inherent uncertainty within the measurements. Uncertainty in element identification can be determined by locating the photopeak centroid at the full peak energy location then determining accuracy verses a known, expected value, shown in a normal distribution. Gamma intensity uncertainty can be determined by using the activity peak width at the full width half max height. A Poisson distribution can be used to show the probability that the measured intensity is within an acceptable level of uncertainty.

Section 3.3 Detection of NaCl

The gamma spectrum resulting from the neutron activation of NaCl (salt) can be seen in Figure 3.5 and 3.6. The spectra do not show defined photopeaks for both the chlorine gamma energies of interest. Analysis shows the counts within the acceptable region of the energies for Cl, 1165 keV and 6619 keV, in both Figures 3.5 and 3.6 is within the standard deviation of the baseline as well as below the minimum detectable limit. Figure 3.7 shows the Cl at 6110 keV, but once again there is no detectable signal for the chlorine at this energy. This may be due to the photopeak of O at 6130 keV engulfing the 6110 keV photopeak, but it did not show any skew towards 6110 keV when compared to other spectra with no chlorine samples. Sodium is also present in NaCl,

however this author was only concerned with the detection of chlorine due to its weaponized nature.
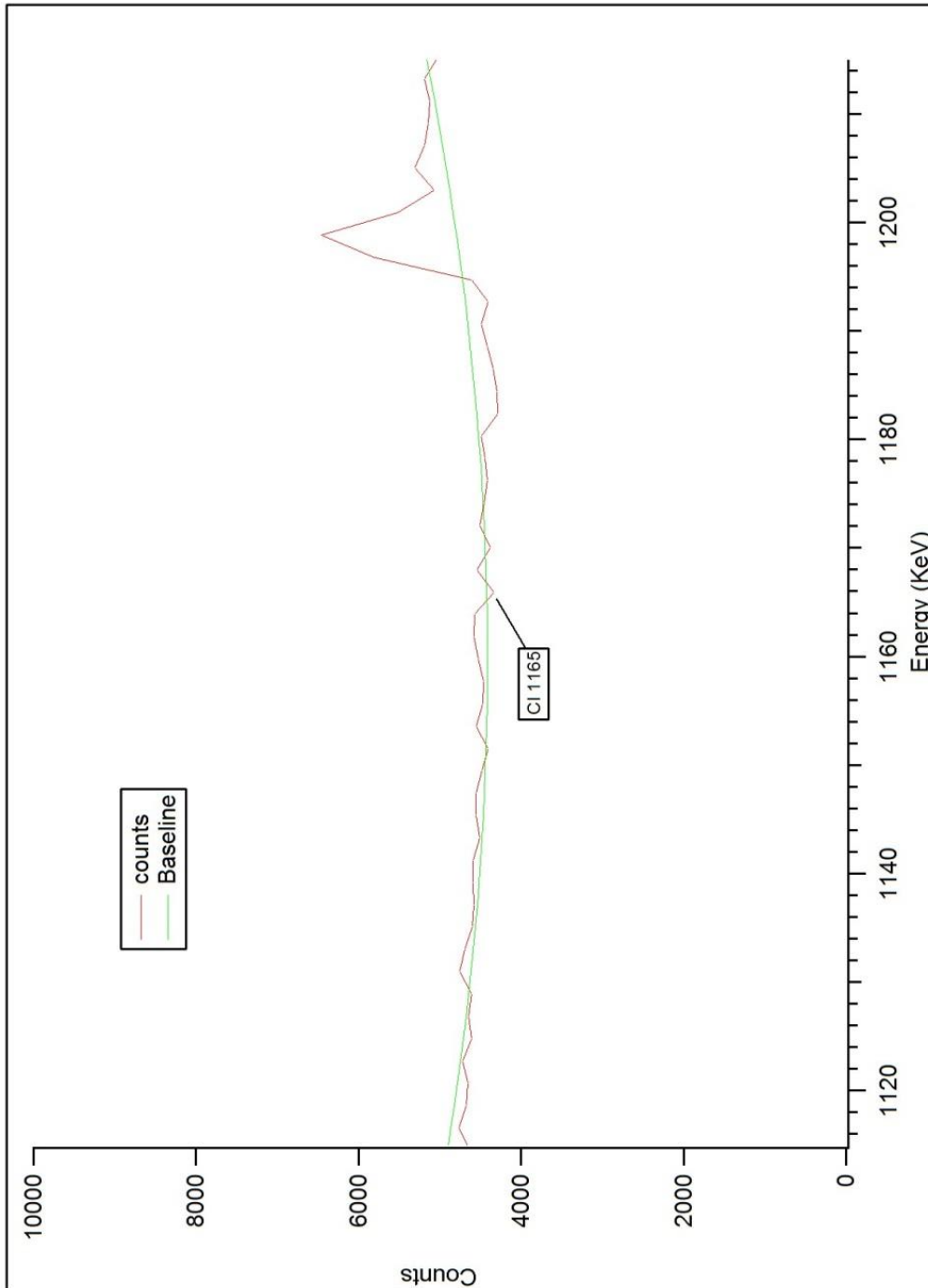

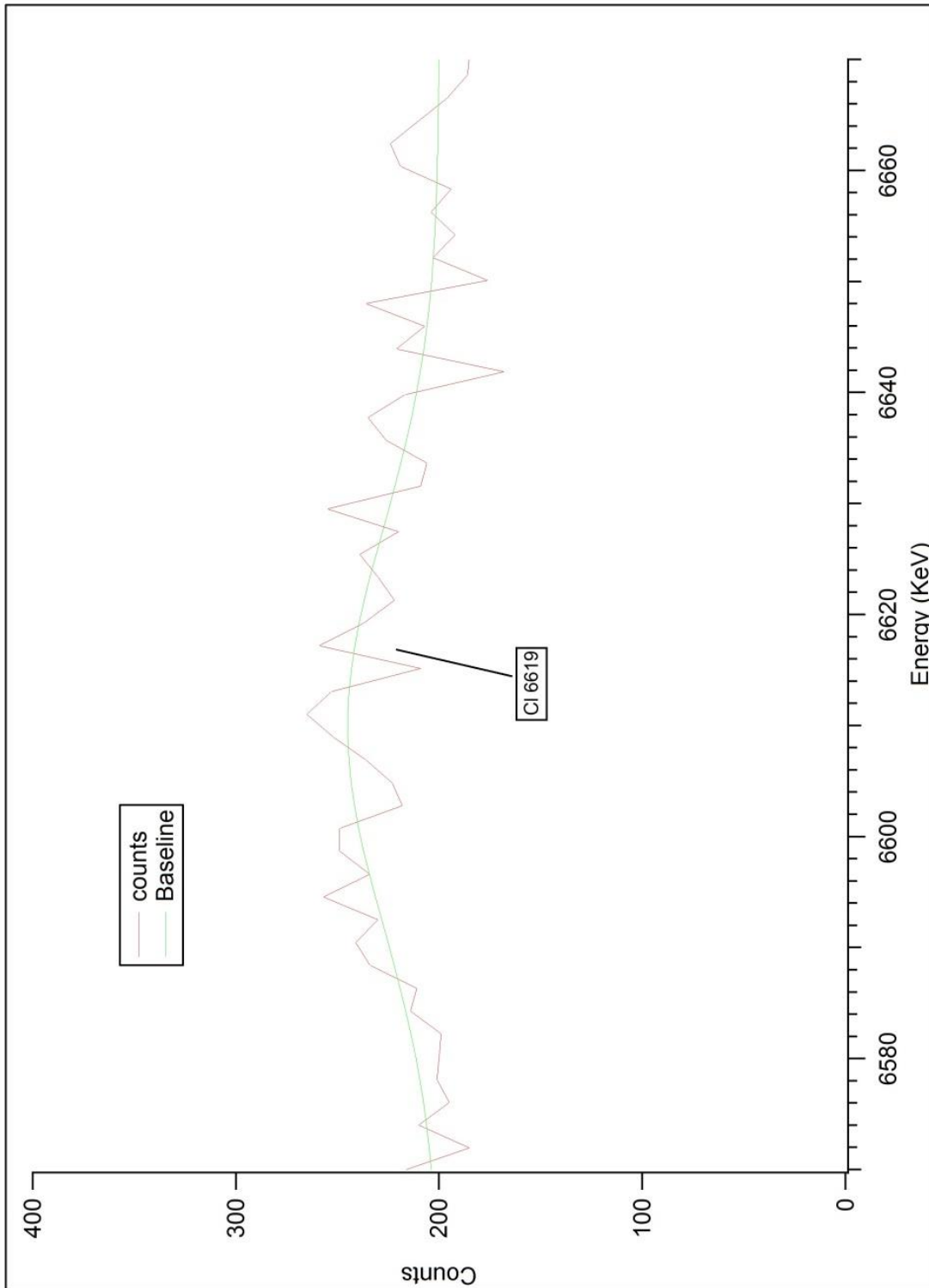**Figure 3.5** Gamma spectrum of neutron activated NaCl underwater at 1165 keV

Figure 3.6 Gamma spectrum of neutron activated NaCl underwater at 6619 keV
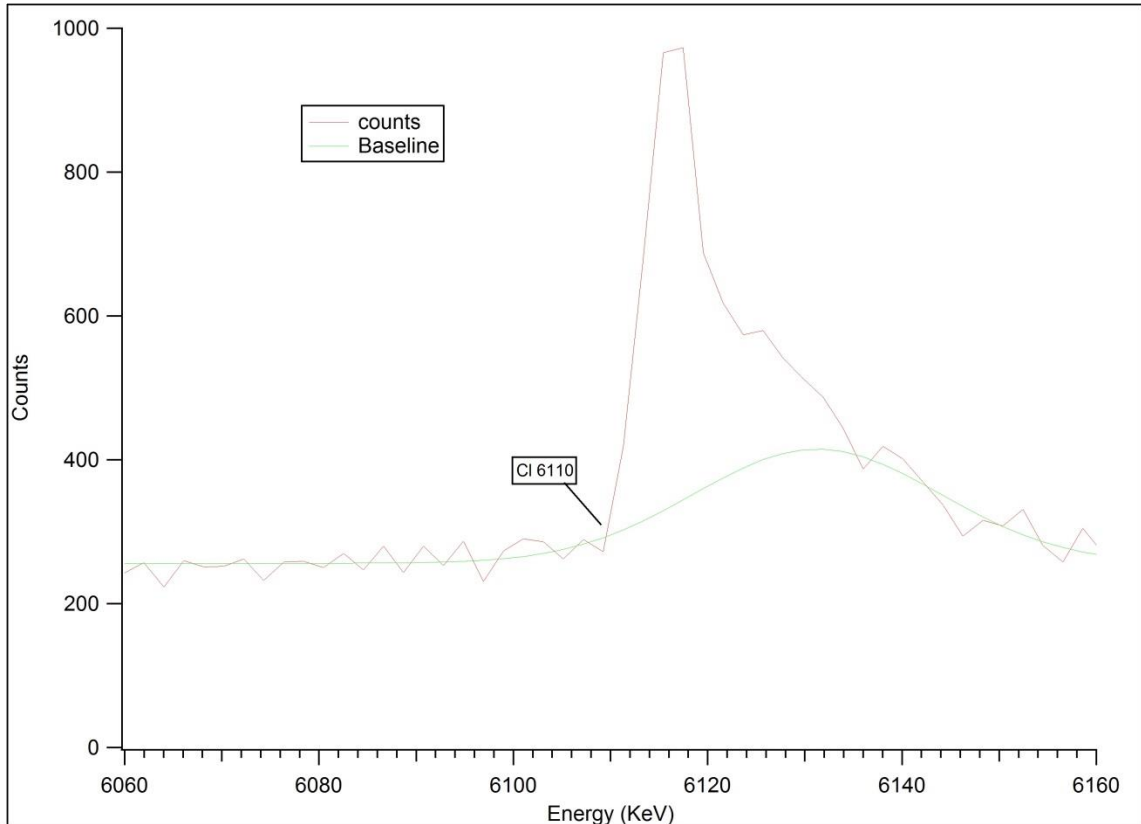
Figure 3.7 Gamma spectrum of neutron activated NaCl underwater at 6110 keV

Section 3.4 Detection of Sulfur

The prompt gamma rays produced by sulfur when it captures thermal neutrons in between the 14 MeV neutron pulses should display a signal at 841 keV and 5420 keV. The signal at 841 keV is, however unuseful due to too much interference. Therefore, this analysis focuses on the energy signal at 5420 keV. This spectrum was once again analyzed as before and it was determined sulfur (S) does not have a detectable photopeak using this methods utilized in this experiment. Figure 3.8 shows that there is no visible photopeak above the standard deviation of the baseline.
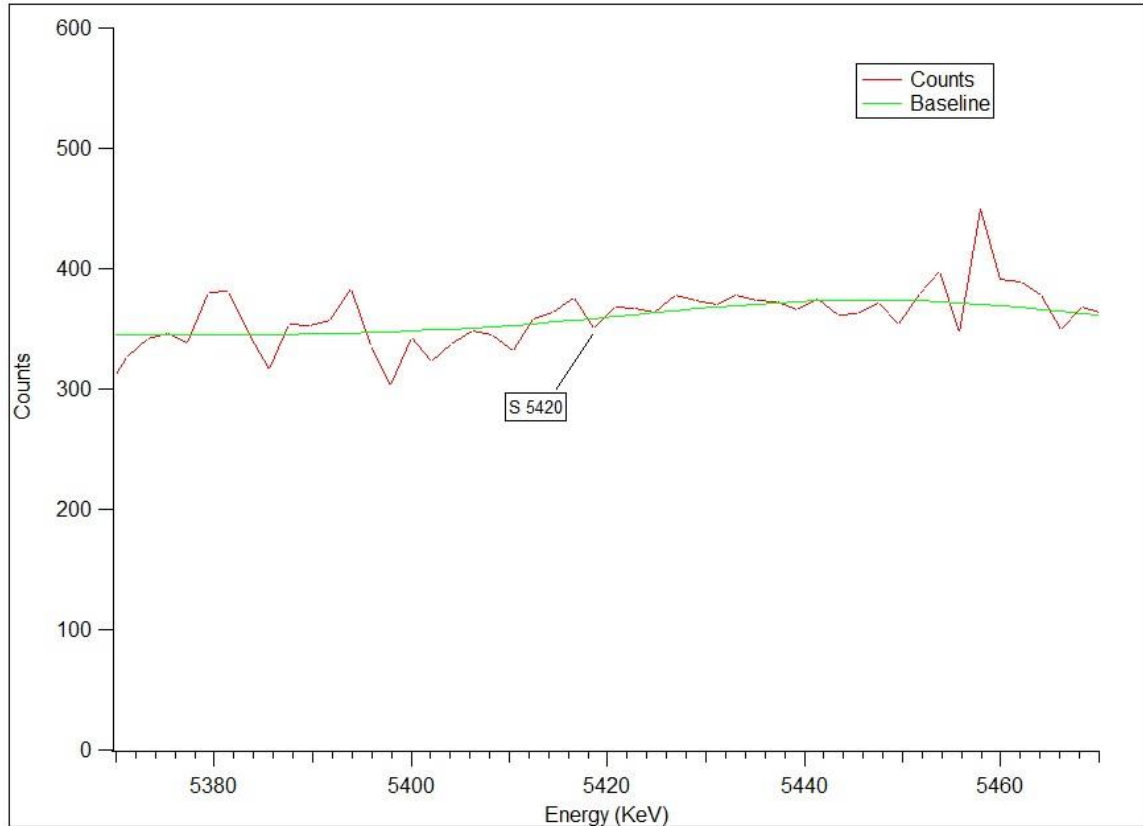
Figure 3.8 Gamma spectrum of neutron activated sulfur at 5420 keV underwater

Section 3.5 Detection of Nitrogen

In this gamma spectrum from the activation of a urea sample in the underwater environment, a photopeak can be observed which could contain the gamma signature of both oxygen second release and that of nitrogen . The photopeak shows that since the gamma signatures of both elements are extremely close together, with oxygen having a characteristic gamma of 5108 keV and N being at 5110 keV, this could be a dual photopeak. In this spectrum, there are not two defined photopeaks at approximately 5110 keV. Although there is a clear valley between the peaks, which could constitute the different signatures of both oxygen and nitrogen, looking at the skew of the centroid of the photopeak when compared to other samples shows that this is not skewed towards a
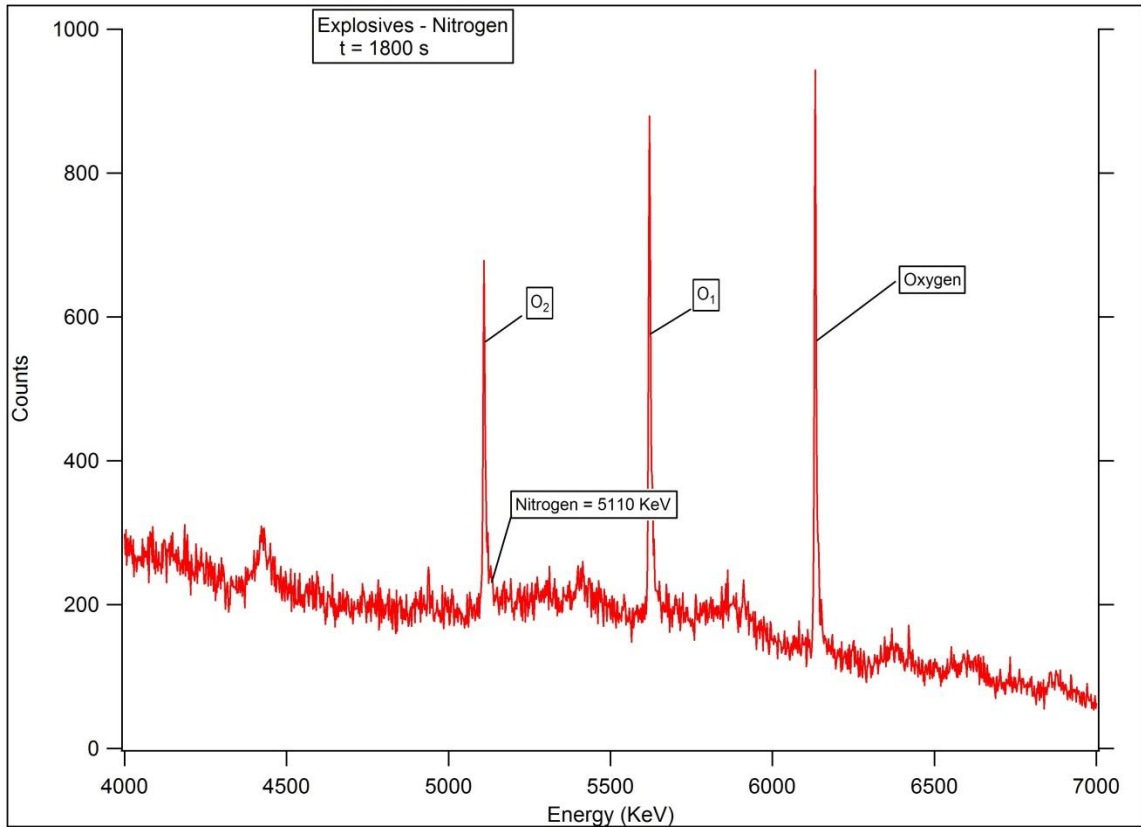
36

nitrogen 5110 keV photopeak.

Figure 3.9 Gamma spectrum of neutron activated nitrogen (5108 keV) underwater

Nitrogen also produces a 10833 keV gamma signature from the (n, γ) reaction. This energy level is towards the maximum detectable energy for the dectector used in this experiment. Analysis of this portion of the spectrum once again shows no detectable photopeak of nitrogen above the minimum detectable limit when determined using standard deviation. Figure 3.10 shows this spectrum in relation to its baseline measurement.

Urea samples can be detected using their C/O and C/N ratios. This experiment showed that the gamma intensities of C and O in the Urea were actually less than that of the gamma intensities of the background measurements when comparing the net

photopeak areas of the two. Table 3.3 shows the net photopeak areas of C and O from the Urea compared to that of the background. These areas were determined by subtracting the baseline from the counts and then calculating the area of the remaining photopeak. Higher concentrations of C and O were expected in the urea sample reading, but were not detected.
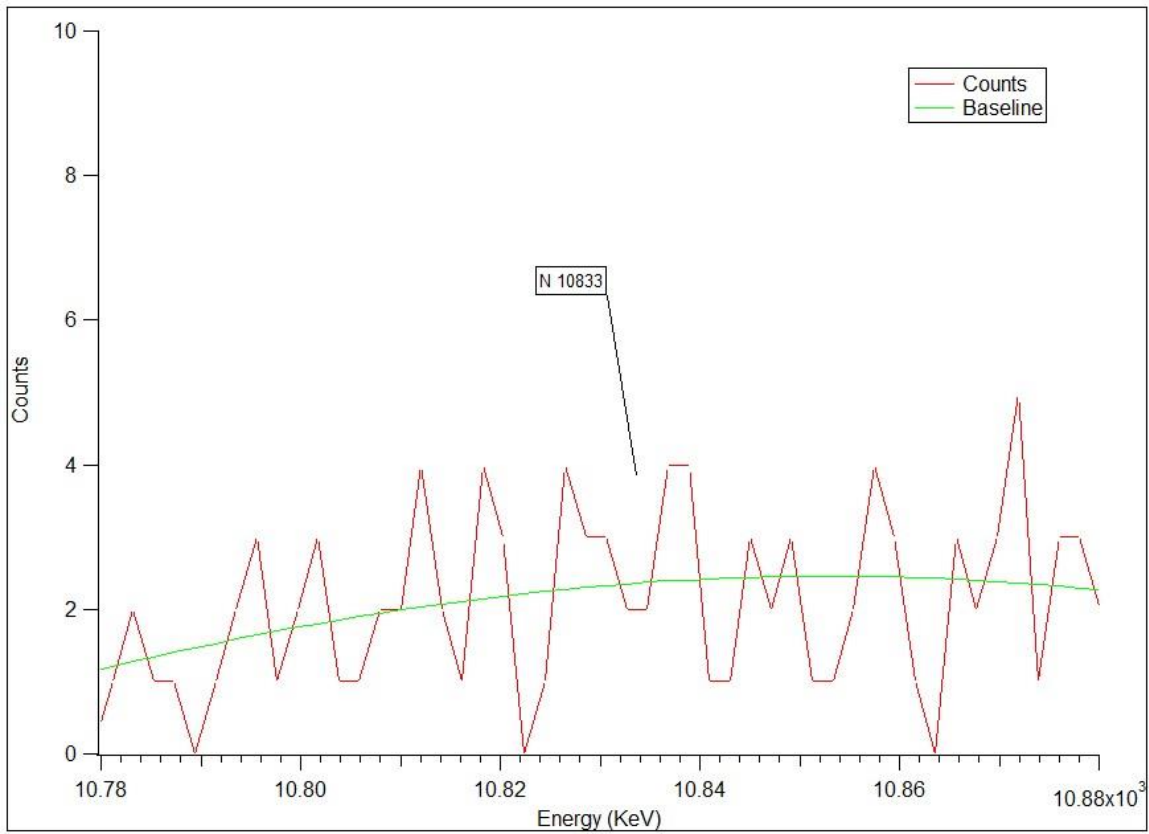

Figure 3.10 Gamma spectrum of neutron activated nitrogen at 10833 keV underwater

PHOTOPEAK AREA		
	BACKGROUND	**UREA**
C	185.33	22.525
O	2347.1	2225.2

Table 3.3 Photopeak areas of C and O in urea and background

Section 3.6 Detection of Narcotics

The gamma ray spectrum resulting from the neutron activation of the cocaine surrogate can be seen in Figure 3.11, Figure 3.12 and Figure 3.13. The major elements to be detected in narcotics are nitrogen (N), carbon (C), oxygen (O) and hydrogen (H). The photopeaks show extreme count levels for both hydrogen and oxygen. Extreme counts can be observed for oxygen in Figure 3.11. This is expected due to the experiment being conducted underwater. Due to the neutron activated hydrogen and oxygen from the water, comparisons to the background gamma intensity for H and O can be used to determine the detectability of the samples. However, in these trials in the background readings the gamma intensities for that of C and O are greater than that of the gamma intensities in the trials measuring the simulant. This is also true for the Urea measurements. This results in a confounding of the readings because there is no baseline to compare the O photopeak to making it impossible to get an accurate C/O or C/N ratio.

An accurate measure of O is required to make a determination of the presence of nitrogen based fertilizer compared to other materials.

Nitrogen is potentially observed as being detected in Figure 3.11, as there is a dual peak visible once again at approximately 5110 keV which is where oxygen and

nitrogen share a charactersitic gamma energy, at 5108 keV and 5110 keV respectively. This dual photopeak once again has a valley seperating the peaks, but analysis of the peak shows no skew towards a N photopeak when compared to other samples. Figure 3.12 examines the presence of N at the 10833 keV photopeak.

The attempt at making any discernable measurement of a carbon photopeak can be viewed in Figure 3.13. In this spectrum carbon is not shown as present as there is no photopeak above the minimum detection limit.
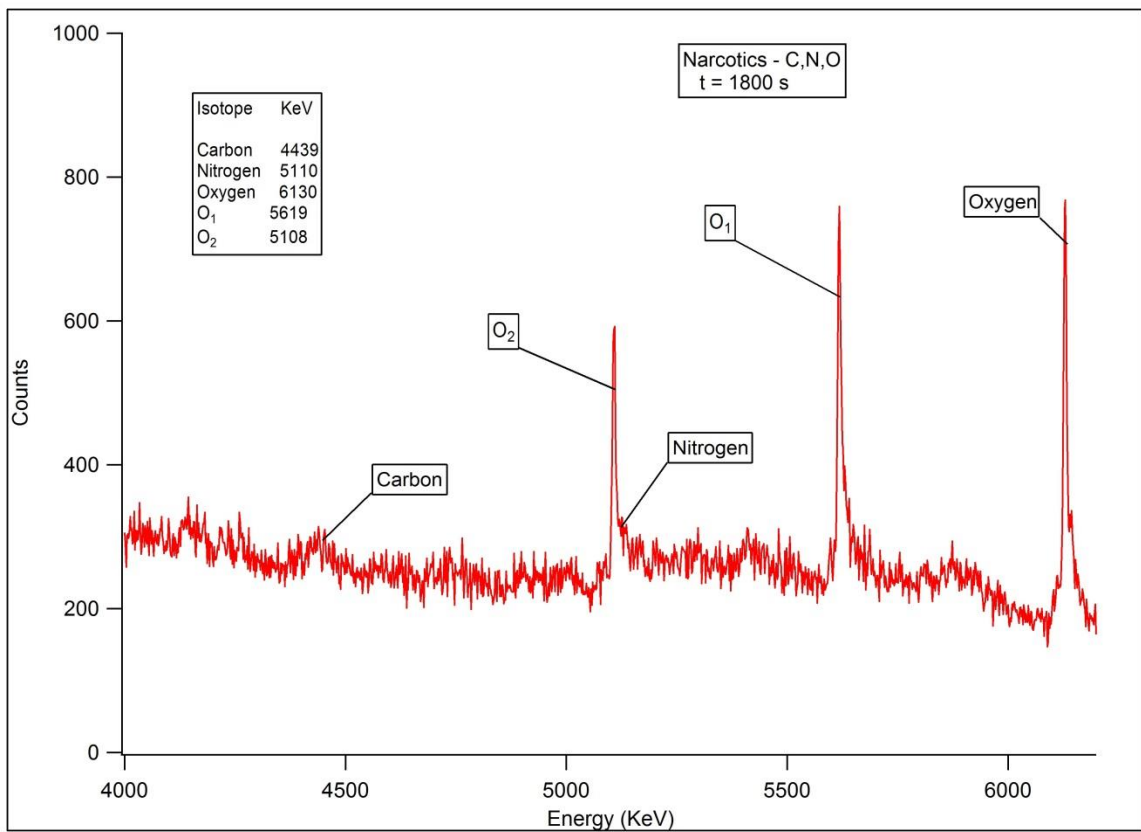

Figure 3.11 Gamma spectrum of neutron activated cocaine surrogate underwater (N at 5110 keV)
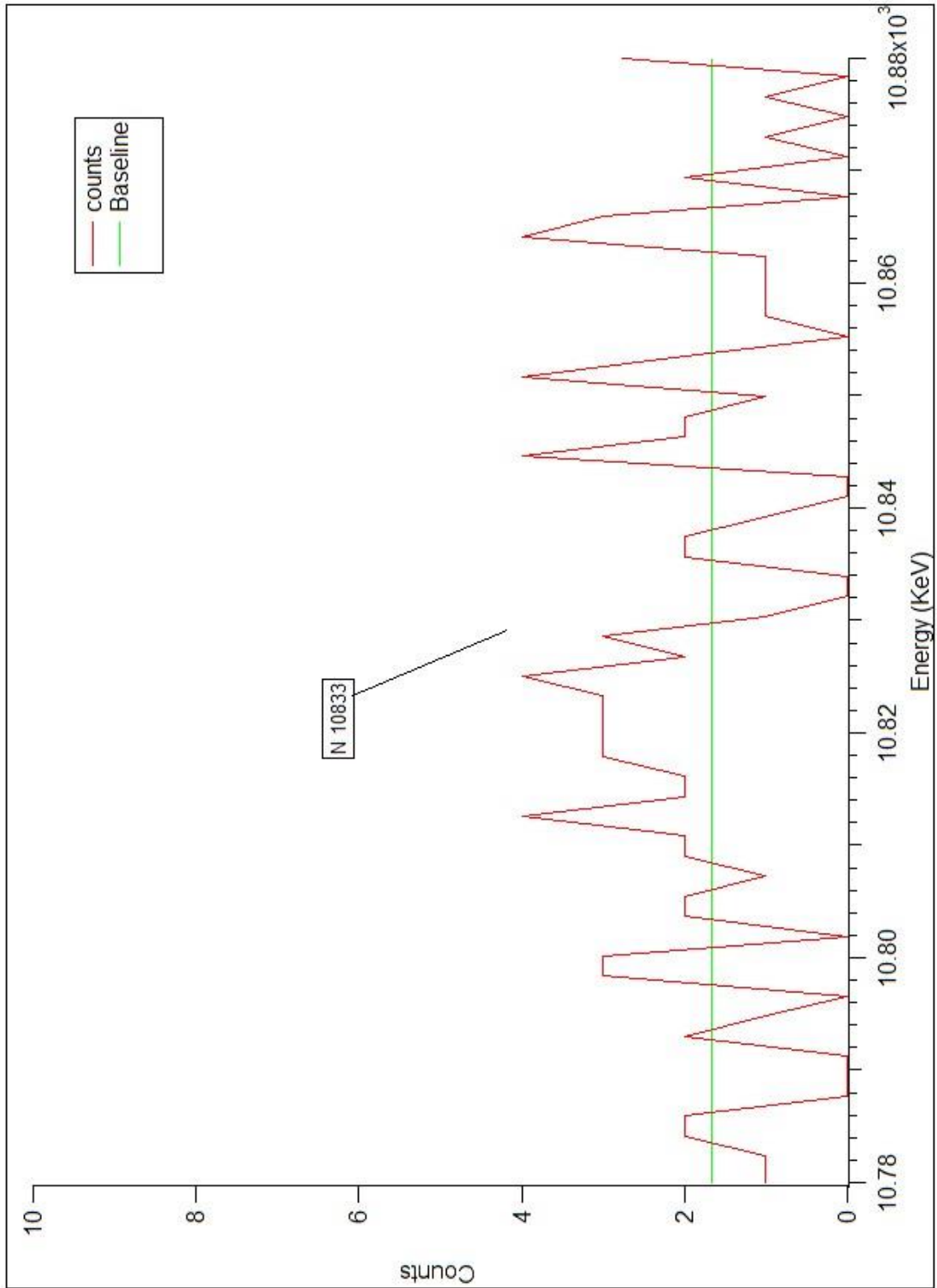
Figure 3.12 Gamma spectrum of neutron activated nitrogen at 10833 keV underwater in
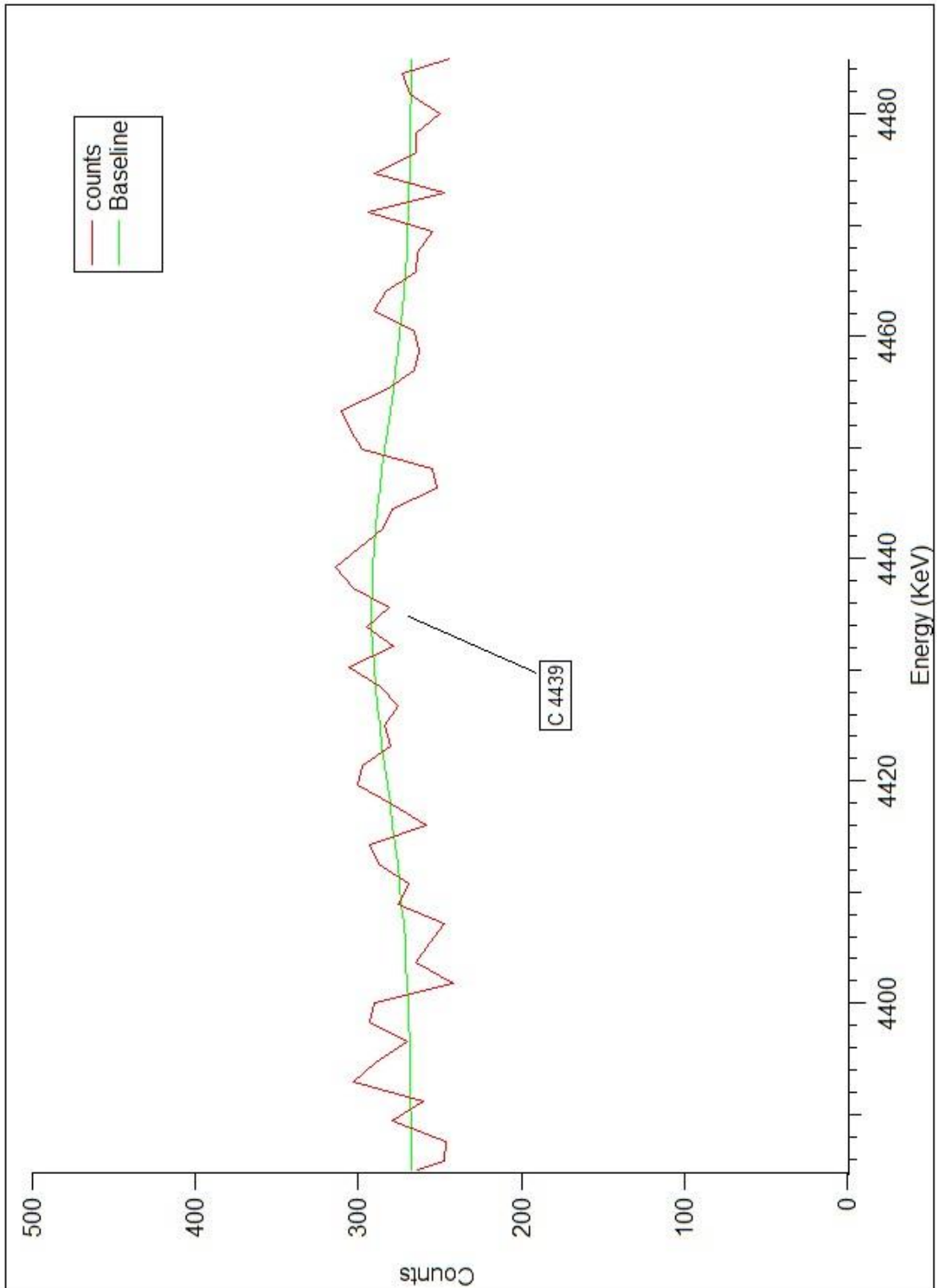
a cocaine simulant

Figure 3.13 Gamma spectrum of neutron activated carbon (4439 keV) from the sample

placed in the underwater enviroment

Section 3.7 Results Overview

The gamma ray spectrum for each sample material shows no detection qualities in the underwater environment. The NAA of NaCl showed no defined photopeaks for either Cl or Na. Chlorine has been measured at observable photopeaks of 1165, 6110 an 6619 keV in previous studies, but did not show a presence in the samples of NaCl when measuring prompt gamma rays. The sample of sulfur which was subject to the same PGNA analysis does not show a defined photopeak at its characteristic gamma energy of 841 keV or at its known prompt gamma energy of 5420 keV.

In regards to nitrogen rich material detection using FNAA from the 14 MeV pulse no discernable data could be recorded for C or N making C/O and C/N ratios impossible. Also, innacurrate measurements of O due to the high concentration of water present in the enviroment further hampered the detection of the nitrogen rich materials.

Chapter 4 – Discussion

The intended goal of this project was to determine the feasibility of using PFTNA in an underwater environment to protect human life from unknown threats that may be looming in ports and waterways. If this technique can be utilized in a practical manner to determine the contents and make up of devices attempted to be concealed underwater, this author believes this technique can protect the lives of the operators that are going hands on with these concealed threats by giving them the foreknowledge of what exactly they will be handling.

It was shown in Barzilov, Novikov and Womble (2009) using Monte Carlo Simulations, that detection of chemical materials underwater using PFTNA and isotropic neutron production is indeed possible. The assembled experimental setup used in this experiment, was tested in relevant environmental conditions. It has been shown that the system can indeed detect gamma rays from the irradiated environment. However, we note that there are a number of difficulties.

- It is difficult to differentiate an oxygen signal from an irradiated sample from the background oxygen signal.
- The water environment acts as a very effective shielding and thermalizer to the 14 MeV neutrons. This leads to a significant decrease in production of the (n,n'γ) reaction, and hence a significant suppression of gamma signal from those reactions, which are necessary for detection of C and O.
- The water also acts gamma ray shielding reducing observed gamma signals even more.

- It seems that "naïve" data analysis routines employed in this study doesn't allow for reliable extraction of signals from gamma ray spectra.

Based on obtained results, the following future research can be proposed:

- Other types of gamma ray detectors should be tested. For example, BGO detectors, although having worse energy resolutions, have much higher efficiency, which could lead to higher observed signal.
- A more sophisticated routine data analysis could be used. For example, it was shown that spectral decomposition technique or wavelet analysis could help to extract small signals in a presence of high background signal.
- Further Monte Carlo based simulations are needed to optimize geometry of the system. Such parameters as a geometrical size of shielding, and different varieties of shielding materials should be optimized more so.

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