Propane as an Alternative Gas in a Constant Flow Proportional Counter

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Abstract

The purpose of this project was to determine if a low cost hydrocarbon gas could be successfully used in a constant flow proportional counter rather than a traditional gas such as P10. Portable propane commonly used for welding or cooking in small disposable containers was chosen to be tested. Four beta sources, carbon-14, technetium-99, strontium-90/yttrium-90, and tritium, were tested in a constant flow proportional counter with both P10 gas as well as propane. The propane gas tests showed a clear increase in count rate as a function of voltage for three of the sources. This supports that there is gas multiplication from propane, and it is possible substitute for P10 gas. The resolution of the spectra however, is not as clear as the P10 gas.

Introduction

A constant-flow proportional counter is a radiation detector capable of detecting both alpha and beta particles. This type of detector is commonly used by health physicists to determine the radioactivity of a small source, or a contaminated sample (Cember and Johnson, 2009). The detector works by having a high voltage electric field within a gas filled chamber in which radiation enters and ionizes the gas inside. The electric field is created by the chamber having a voltage bias between the inner wall and a thin wire in the center of the chamber. Ionization from a radioactive particle creates electrons which, when in a strong enough electric field, collide with other molecules, creating a cascade of electrons moving toward an anode, or wire, within the chamber (Tsoulfanidis, 1995). When the electrons reach the anode, a voltage signal will be registered as a count. Higher energy ionizing particles produce more electrons, thus creating a higher amplitude voltage signal. The pulse height from each signal can determine which voltage range should be used for each detector type. Figure 1 from *Introduction to Health Physics* by Cember and Johnson displays pulse height versus voltage, illustrating the relative regions of optimal voltage used for each detector type. A proportional counter must be used at voltages which correspond to these regions.

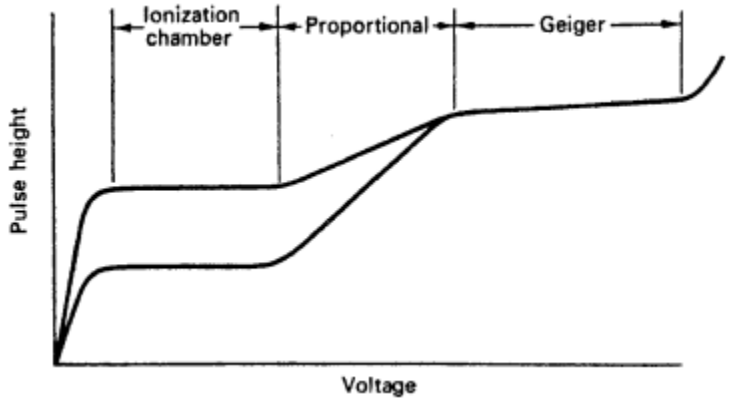

Figure 1. Pulse height vs voltage in a proportional counter. (Reprinted from Cember, H. and Johnson T. E., 2009, *Introduction to Health Physics,* 4th edition, McGraw-Hill.)

The liberation of electrons from one original ionization is known as gas multiplication. This is fundamental to the operation of a proportional counter. Standard air does not have the property of gas multiplication; thus, an alternative gas, capable of multiplication, must be pushed into the proportional counting ion chamber. A common gas used in proportional counters is P10 which consists of 10% methane (CH4) and 90% argon (Ar). Argon is the main gas which is ionized by radiation, sending ions to the cathode, and electrons to the anode. When ions of the main gas reach the cathode, or outer wall, they can eject another electron, creating a separate signal not caused by the radiation. Methane is known as a quenching gas because the molecules dissociate when hitting the cathode, rather than create extra electrons. These methane molecules can also ionize when colliding with an argon ion. This does not allow as many argon ions to reach the cathode, resulting in fewer false signals (Tsoulfanidis, 1995). As voltage increases, gas multiplication increases. This can be seen by plotting gas multiplication vs voltage, as displayed in Figure 2 (Cember and Johnson, 2009).

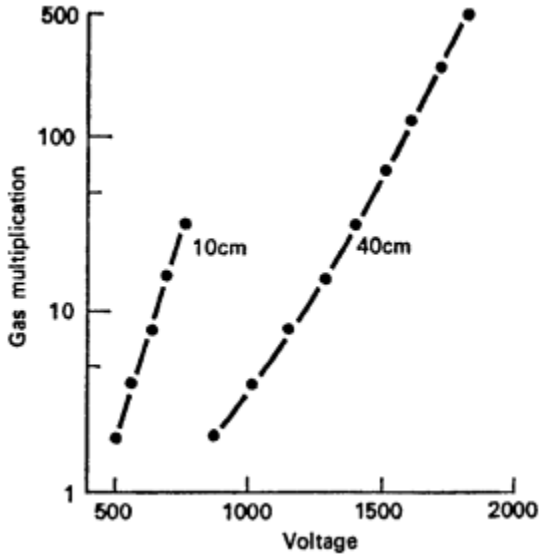

Figure 2. Gas multiplication vs voltage for two different pressure argon filled proportional counters. (Reprinted from Cember, H. and Johnson T. E., 2009, *Introduction to Health Physics,* 4th edition, McGraw-Hill.)

Excessive voltages produce overarcing, which results in false counts. These false counts are generated when electrons with too high of an energy cause an unending cascade Tsoulfanidis, 1995).This occurs when the voltage passes the right end of the proportional region in Figure 1. The region where high voltage works in proportional counters is the high voltage plateau seen in Figure 3 (Tsoulfanidis, 1995). Beta Particles and Alpha particles have their own respective plateau in a count rate vs voltage as seen in Figure 4 (Tsoulfanidis, 1995). When looking for a beta particle, the voltages to the left of the plateau will have no count rate, and to the right of the plateau the signal will drastically increase from overarcing (Tsoulfanidis, 1995).

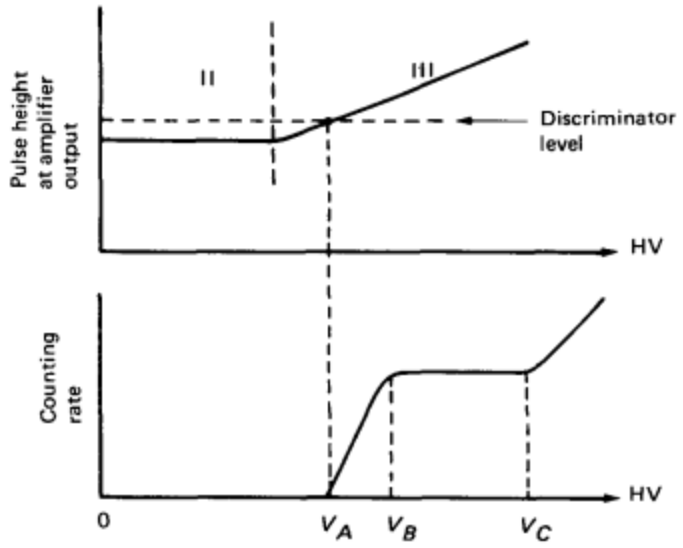

Figure 3. High voltage plateau (lower curve) in counting rate vs voltage plot. (Reprinted from Tsoulfanidis, N., 1995, *Measurement and Detection of Radiation,* 2nd edition, Taylor & Francis, Washington D.C.)

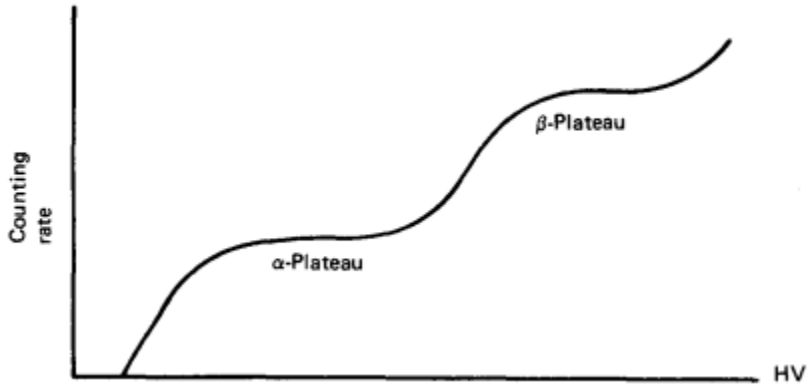

Figure 4. Alpha and Beta plateaus from a proportional counter. (Reprinted from Tsoulfanidis, N., 1995, *Measurement and Detection of Radiation,* 2nd edition, Taylor & Francis, Washington D.C.)

The strength of signal from the ion chamber depends on how many electrons reach the anode after the gas multiplication of the original ionization. Particles with more energy can ionize multiple gas particles as it passes through the chamber, resulting in a higher amplitude from the higher volume of electrons reaching the anode in a short time frame. These strength of signals can be discriminated in order to only see counts from particles of certain energies. The particle's signal is small relative to the voltage of the chamber. In order to see and differentiate signals, the original pulse needs to be amplified by a large amount. A preamplifier is used to adjust the signal from the detector and increase it to be noticeably different from electronic noise

(Tsoulfanidis, 1995). An amplifier is then used to increase the signal by a large factor, making it possible to differentiate voltage pulse heights (Tsoulfanidis, 1995).

A single channel analyzer can then determine a range of pulse amplitudes that will be counted. This can also be known as a discriminator. It helps to reject noise or other unwanted pulses from being counted.

A coincidence is a second discriminator in a sense. It is implemented when there are two proportional counters in use. The coincidence works by rejecting two similar pulses within a short window of time. This is done when two proportional counters are stacked with the source being in between the two. A high enough energy particle could come from outside and pass through the two counters leaving two pulses not from the source. The coincidence will check for similar pulse amplitudes, if they occur within a set timeframe (within nano or microseconds) the pulses will not be counted. Cosmic rays or radioactive sources outside the detecting area are examples of signals the coincidence can eliminate.

A linear gate can be used with a coincidence and is what allows a signal produced from the coincidence to reach the multichannel analyzer. If the coincidence does not detect a double pulse, it sends the count to the linear gate. The gate then opens for that signal and registers it to the multichannel analyzer.

A Multichannel Analyzer (MCA) records the pulses according to their amplitude. Since the amplitude is proportional to the energy of the particle that produced it, each pulse height has a respective channel it is stored in. The spectrum of a source can be seen after a test with the MCA showing a histogram with counts vs channel number.

If the proportional counter detects gas multiplication from propane, indicated by an increase in count rate vs voltage, then propane is a suitable replacement for a more traditional gas. A basic proportional counter is shown in Figure 5. For more on proportional counters see *Measurement and Detection of Radiation* by Tsoulfanidis.

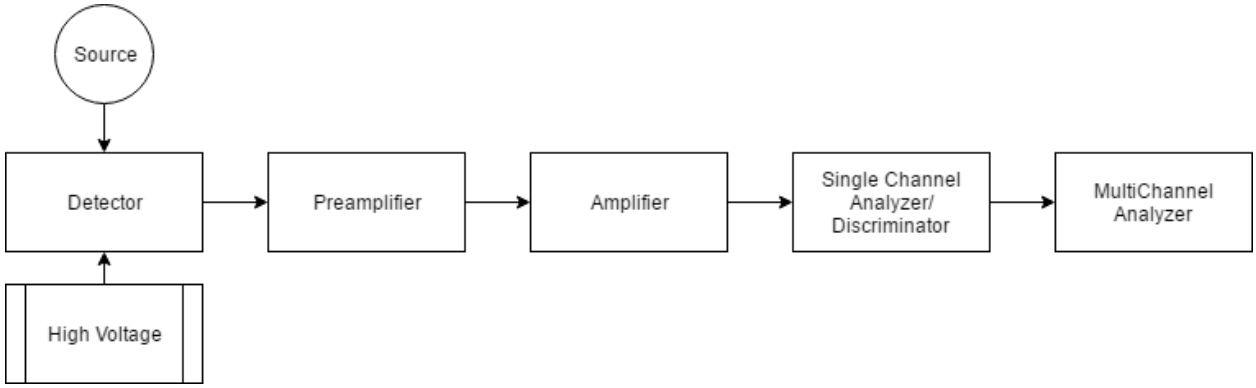

Figure 5. Schematic of a basic proportional counter setup.

Methods

For this experiment, the following equipment was used:

- P10 Gas
- Bernzomatic Propane 14.1 oz
- Proportional counter
- Lead shielding
- Preamplifier
- Tennelec FM 40A Gas Flowmeter
- Ortec 426 Linear Gate
- Ortec 427a Delay Amplifier
- Canberra 2040 Coincidence
- Ortec 551 Timing SCA
- Ortec 575A Amplifier
- Ortec 550 SCA
- Canberra 2012 Amplifier
- Ortec 556 High voltage supply
- Genie 2000 Multichannel Analyzer
- Canberra 2000 NIM bin power supply
- Advantech 610 Industrial Computer
- Brass high pressure valves
- Pressure regulator with brass connections
- Gas flow regulator with brass connections

The setup of this equipment is shown in Figure 6. Table 1 shows the settings for components that were constant throughout the duration of the experiment.

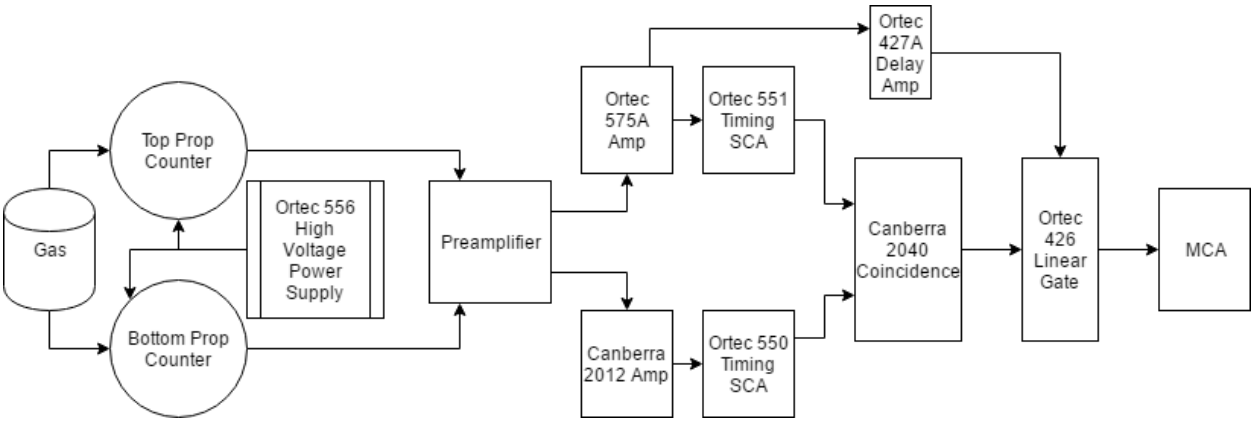

Figure 6. Schematic of the proportional counter used in this experiment.

NIM Bin Components					
Ortec 426 Linear Gate	Ortec 427A Delay Amp	Canberra 2040 Coincidence	Ortec 551 Timing SCA	Ortec 550 SCA	Canberra 2012 Amp
Norm	.25 µsec (delay in)	10 (resolving time)	0 (Upper level)	0 (Upper Level)	8 (Course gain)
	0.5 µsec (delay out)	0.1-1 µsec (range)	0 (Lower level)	2 (Lower level)	0 (Fine gain)
	1 µsec (delay out)		1 (Delay)	Integral (Switch)	+ (Input Polarity switch)
	1 µsec (delay in)		0.1-1.1 µsec (Walk Adj switch)		
	1 µsec (delay in)		INT (Walk Adj switch)		

Table 1. Constant settings for NIM bin components of proportional counter.

The first step in this experiment was to record beta spectrum of various sources with the P10 gas being used in the detector. The voltage used was 1500 Volts and the gas was flowing with just enough to raise the flowmeter above the bottomed out position. Ten minutes was allowed for the gas to flow into the detector before any tests were done. Four different beta sources were tested. Three were manufactured button sources, and one was a tritium gun sight. The known specifics for each source are shown in Table 2. The Ortec 575A amplifier gain settings, as well as discriminator settings in the Genie 2000 MCA program were needed to be altered for each beta source tested. In order to find the optimal settings for each source, tests ranging from five to ten minutes were done while systematically increasing or lowering the amplifier's course and fine gain settings until the entire energy spectrum could be seen. The MCA program's lower level discriminator (LLD) settings were adjusted for each source to eliminate noise from the lower end of the spectrum. Calculated beta-ray spectra from *A short atlas of beta-ray spectra* by Cross, Ing, and Freedman, as well as *The Beta Ray Spectrum and the Average Beta Energy of Several Isotopes of Interest in Medicine and Biology* by Mantel were used to help determine how each beta spectrum should be shaped. These graphs are theoretical, but are useful in determining noise or overarcing from beta source signals. Figure 7a through 7c show the theoretical spectra for tritium, carbon-14, and strontium-90/yttrium-90. Table 3 shows the optimal Amplifier gain and MCA settings used for each spectra test. A four hour source test, and a 24 hour background was taken for each source with their respective optimal settings. After these tests a count vs

channel number was plotted for each source. The shapes of these spectra would be the basis on what to look for in later tests when propane would be the gas used.

Name	C-14	Tc-99	Sr-90/Y-90	H-3
Activity (μCi)	0.1023	0.01019	0.01025	Unknown
Reference Date	15 May 09	15 May 09	15 May 09	Unknown
Source Number	1369-71-1	1369-71-1	1369-71-1	Unknown
Manufacturor	Eckert & Ziegler	Eckert & Ziegler	Eckert & Ziegler	Unknown
Average Energy (keV)	49.3 (Mantel,1972)	85.4 (Eckert & Ziegler)	196.1/933.5 (Mantel,1972)	5.7 (Mantel,1972)
Maximum Energy (keV)	156 (Mantel,1972)	293.6 (Eckert & Ziegler)	544/2270 (Mantel,1972)	18.6 (Mantel,1972)

Table 2. Information of beta sources tested.

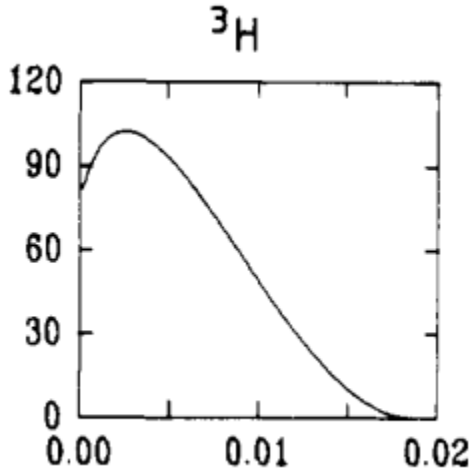

Figure 7a. Calculated spectrum for tritium. (Reprinted from Cross, W.G., Ing, H., Freedman, N., 1983, A short atlas of beta-ray spectra, *Phys. Med. Biol.,* Vol. 28, No. 11, p. 1251-1260.)

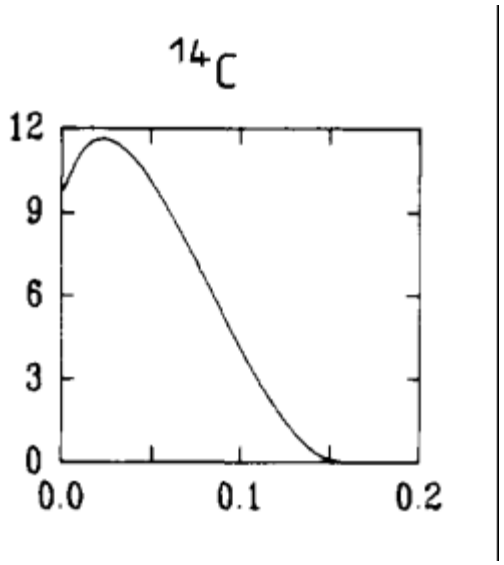

Figure 7b. Calculated spectrum for carbon-14. (Reprinted from Cross, W.G., Ing, H., Freedman, N., 1983, A short atlas of beta-ray spectra, *Phys. Med. Biol.,* Vol. 28, No. 11, p. 1251-1260.)

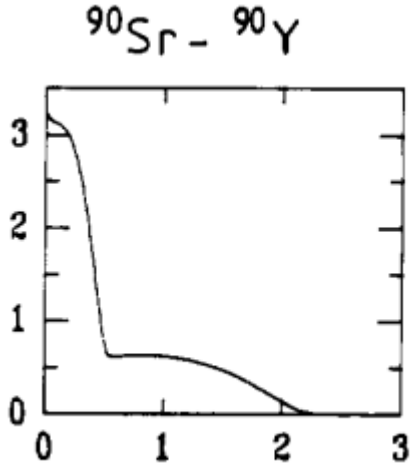

Figure 7c. Calculated spectrum for strontium-90/yttrium-90. (Reprinted from Cross, W.G., Ing, H., Freedman, N., 1983, A short atlas of beta-ray spectra, *Phys. Med. Biol.,* Vol. 28, No. 11, p. 1251-1260.)

Source	C-14	Tc-99	Sr-90/Y-90	H-3
Genie 2000 MCA settings				
LLD threshold %	0.40%	0.40%	0.40%	0.40%
LLD %	3.53%	7.06%	0.00%	6.67%
ULD %	101.37%	101.37%	101.37%	101.37%
Amplifier settings				
Coarse Gain	40	40	2	100
Fine Gain	4.0	4.7	6.0	5.8

Table 3. Ortec 575A Amplifier and MCA settings during P10 gas tests.

The next step was to replace the P10 gas with propane. The propane used was purchased in a portable container at a local hardware store. Brass piping was used to create a valve with an attached flowmeter. The apparatus was connected to the proportional counter with the propane flowing just enough to raise the flowmeter above the bottomed out position. Ten minutes was allowed for gas to flow into the detector before any tests were done. Five to ten minute tests were done while adjusting the Ortec 575A amplifier gain and MCA discriminator settings in order to find optimal spectrum settings for each source. If a spectrum was unable to be seen at the initial 1500 volts, the voltage was increased in increments of 100, while still adjusting the amplifier gain and MCA discriminator settings during five to ten minute tests. Table 4 shows the optimal amplifier gain and MCA settings for the propane tests. A ten minute source test, and a ten minute background were done from for a range of voltages in increments of 100 volts. The range to be tested was determined from a higher voltage where overarcing was apparent, down to a lower voltage where the spectrum was beginning to become difficult to differentiate from electronic noise. The range used for Carbon 14, Technetium 99, and SR 90/y90 was 1500 to 2200 volts, and for Tritium was 1900 to 2200 volts. The tests were done in order from high voltage to low voltage so the discriminator settings would eliminate noise at the high voltages, as well as low voltages. The count rates of each test were calculated and plotted in a count rate vs voltage graph.

Source	C-14	Tc-99	Sr-90/Y-90	H-3
Genie 2000 MCA settings				
LLD threshold %	0.40%	0.40%	0.40%	0.40%
LLD %	3.14%	3.14%	3.14%	5.49%
ULD %	101.37%	101.37%	101.37%	101.37%
Amplifier settings				
Coarse Gain	100	100	100	100
Fine Gain	4.8	4.8	4.8	12.5

Table 4. Ortec 575A Amplifier and MCA settings during propane tests.

Results

Figures 8a through 8d show the spectrum of each source while using the P10 gas at 1500 volts. Through amplifier gain manipulation, entire energy peaks could be seen for all spectra except for Sr-90/Y-90. Comparing to the calculated spectra from Figure 7a through 7c, the carbon-14 and tritium follow a similar bell shaped fall off. The strontium-90/yttrium-90 curve did not follow exactly but does have a noticeable count difference as the channel number increases much like the calculated plot. Compared to the spectra from *The Beta Ray Spectrum and the Average Beta Energy of Several Isotopes of Interest in Medicine and Biology,* again carbon-14 and tritium follow a bell shaped curve. Strontium-90/yttrium-90 could not be compared to this paper as there is no combined spectrum as in the previous paper. Technitium-99 did not have a calculated spectra in either paper and cannot be compared.

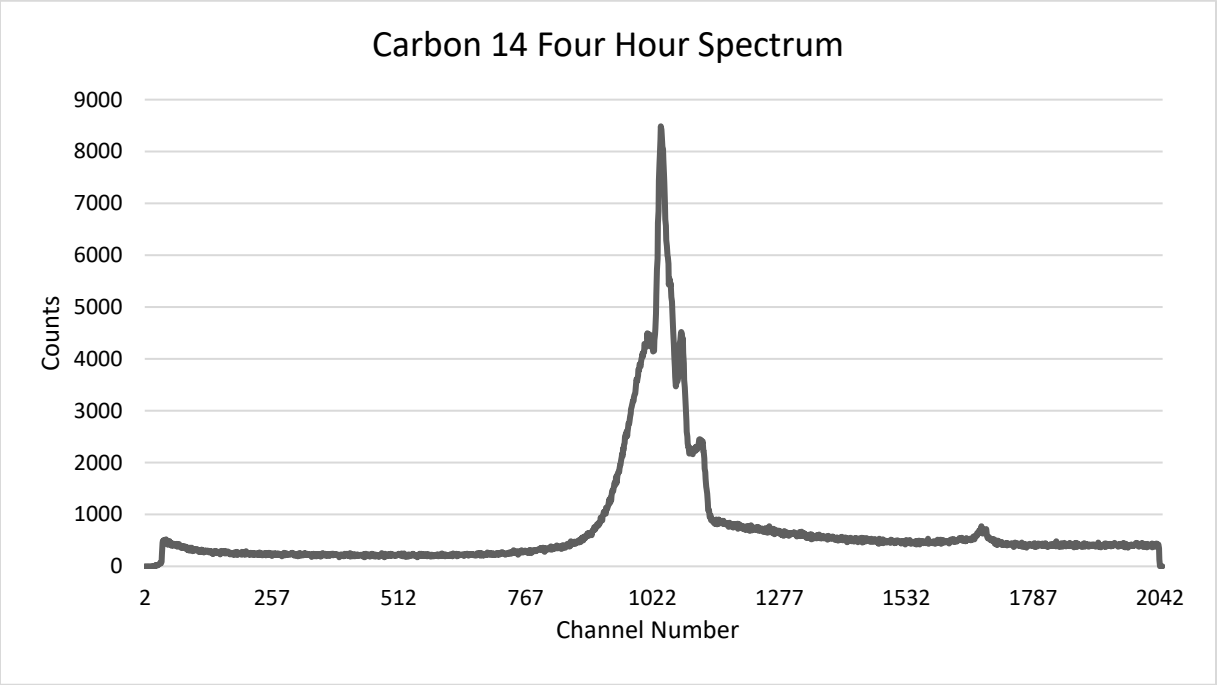

Figure 8a. Carbon 14 spectrum with P10 gas.

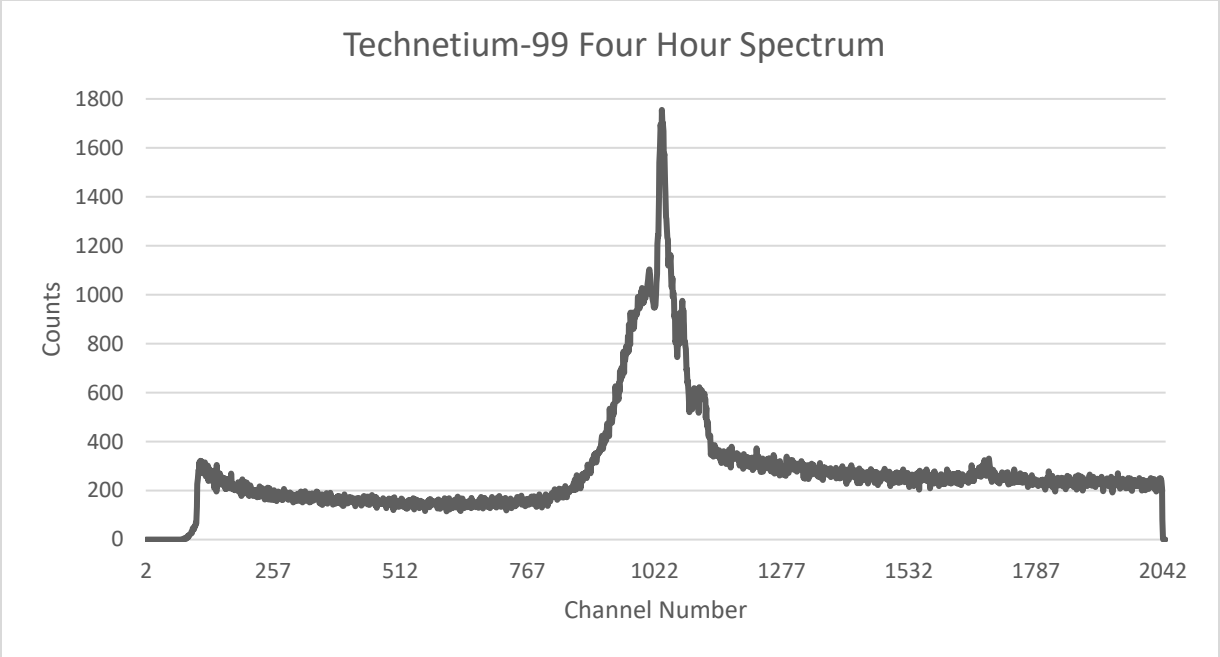

Figure 8b. Technetium-99 Spectrum with P10 gas.

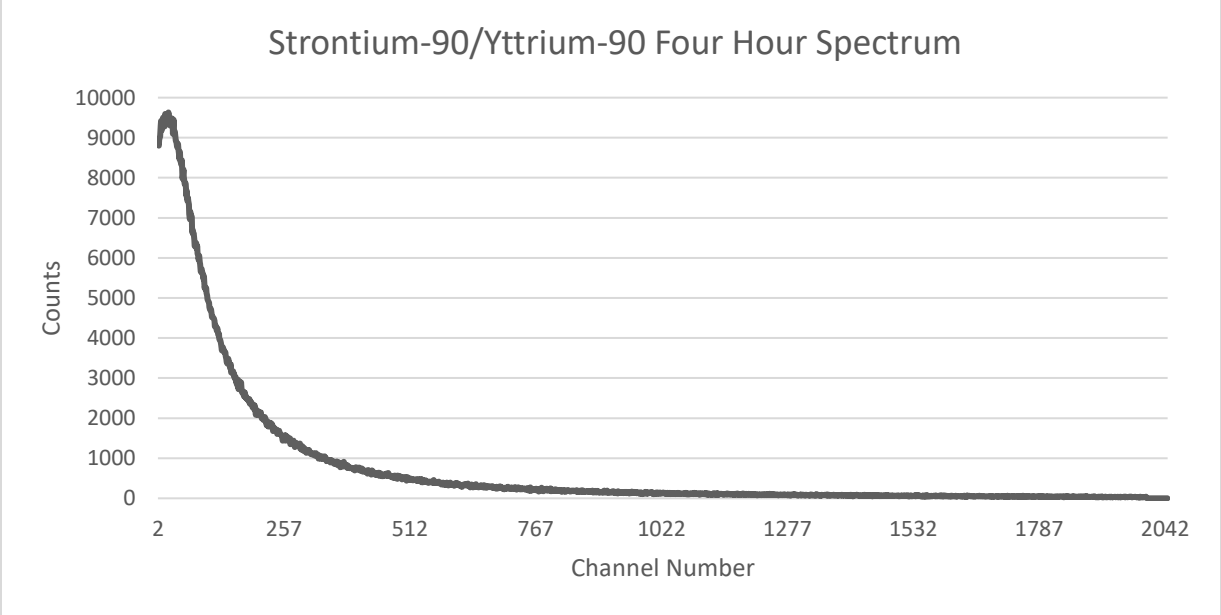

Figure 8c. Strontium-90/Yttrium-90 Spectrum with P10 gas.

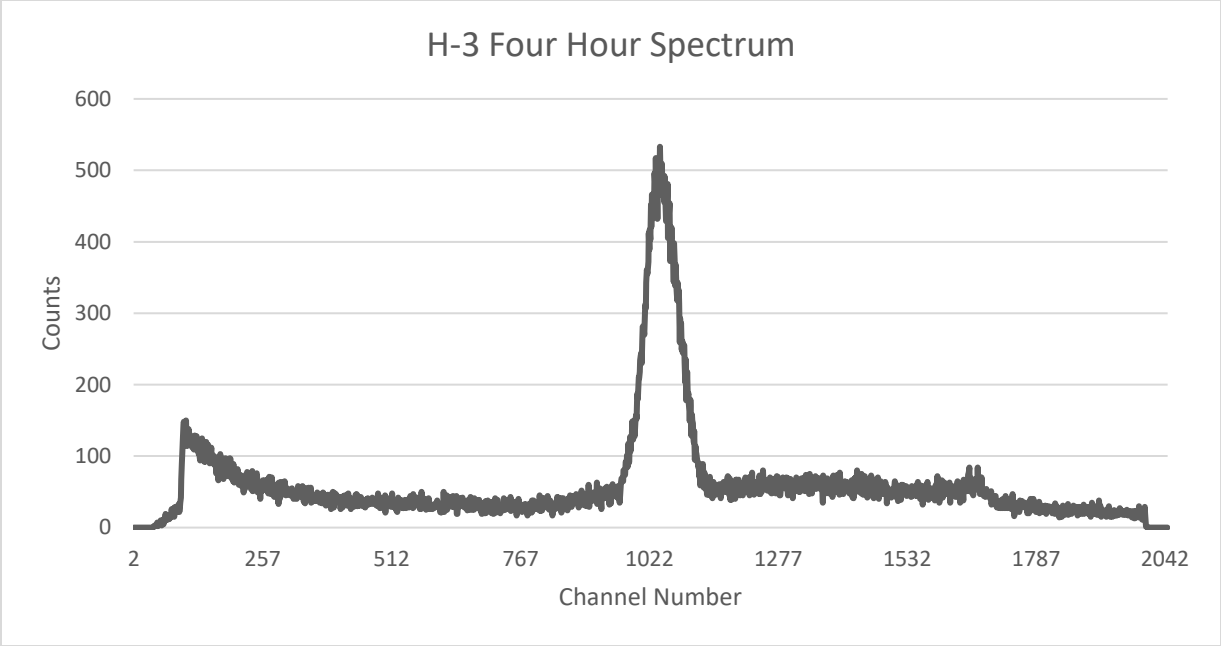

Figure 8d. Tritium Spectrum with P10 gas.

The spectra for sources in the propane test showed similar trends in appearance in comparison to one another. Lower voltages showed what appeared to be the far right of the curves, not showing the peak of the spectra. As the voltage increased, the spectra shifted further right. This is because the pulse height increases with voltage as shown in Figure 1, making the highest energy beta rays create larger pulse heights. Larger pulse heights will be registered at higher channel numbers. Figure 9 and Figure 10 show the differences in carbon 14 spectra at 1500 volts and 1800 volts respectively. It can be seen that this is the far right of the beta ray specrum.

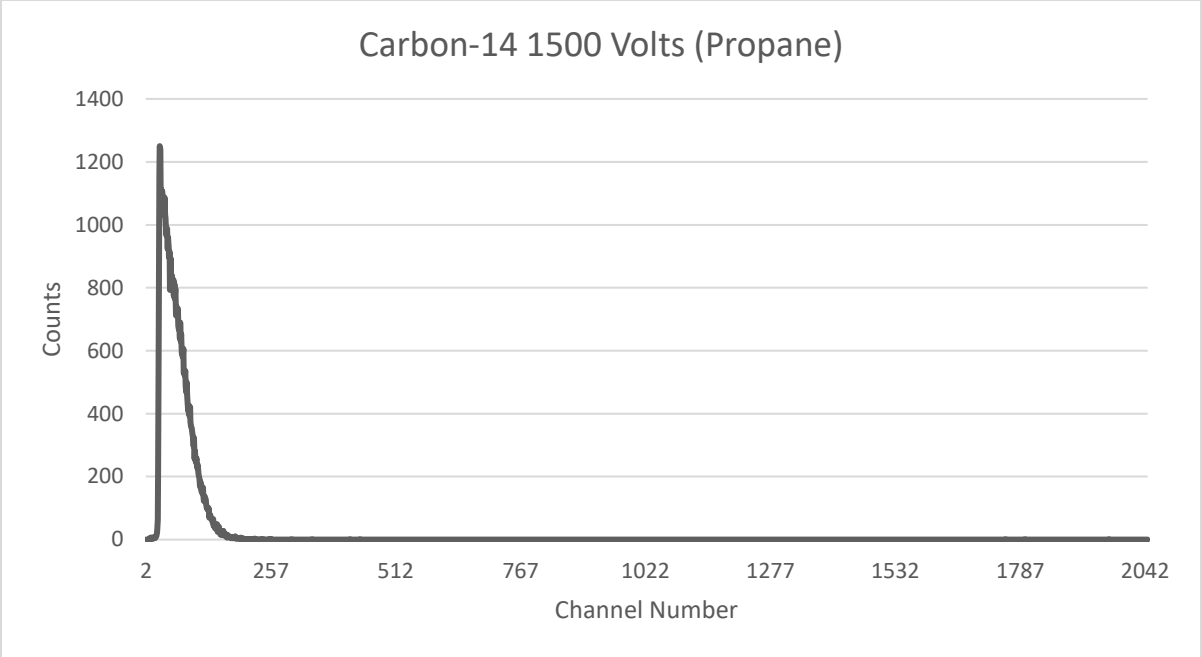

Figure 9. Carbon-14 spectrum at 1500 volts.

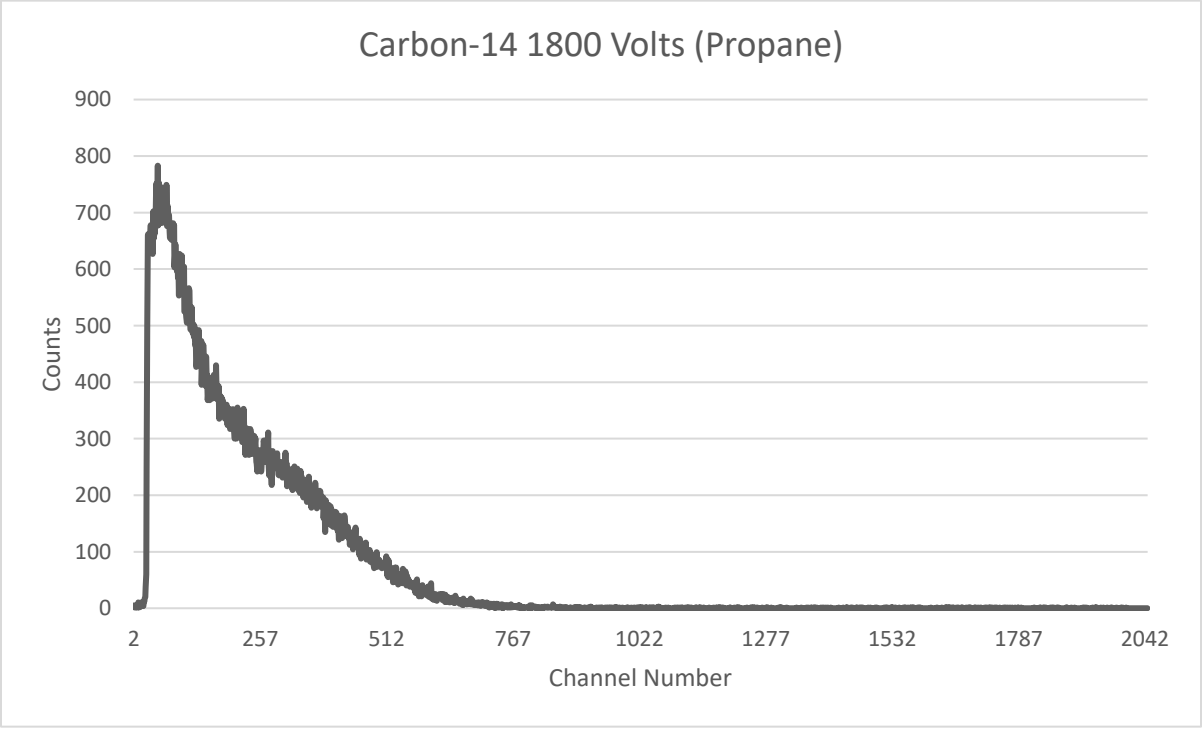

Figure 10. Carbon-14 spectrum at 1800 volts.

Figure 11 shows the results from the propane tests for each source. As shown in the graph, there is an upward trend in count rate for carbon-14, strontium-90/yttrium-90, and technetium-99 until around 2000 volts. Tritium's count rate also decreased at 2000 volts, but so

small it is within the margin of uncertainty. This decrease is caused from overarcing in the background. The background readings began to spike randomly causing the data trends after 2000 volts to be unreliable. An example of this overarcing can be seen in the background test for technetium-99 at 2100 volts shown in Figure 12. This shows that all data past 2000 volts should be disregarded since one interaction can cause multiple false signals. However before 2000 volts, the data points for all sources except tritium have an upward trend in count rate. The region from 1500 to 2000 volts is the high voltage plateau shown in Figure 3. Before this 1500 range the count rate begins to drop off drastically from the signal not even being shown in the spectrum. Tritium however seems to have too small of an energy to create a good beta ray spectrum in this voltage range. Gas multiplication tests include more variables that were beyond the scope of this experiment. However from the high voltage plateau being found, and evidence of overarcing, it appears that gas multiplication is happening. The spectrum is not as clear as the P10 gas because not enough amplification is present. This is likely because the particles do not ionized the propane as easily. Propane appears to be a gas multiplier and could be used in a proportional counter for certain sources. Tritium had too low of energy in this experiment to determine if propane was an adequate gas for low energy beta rays.

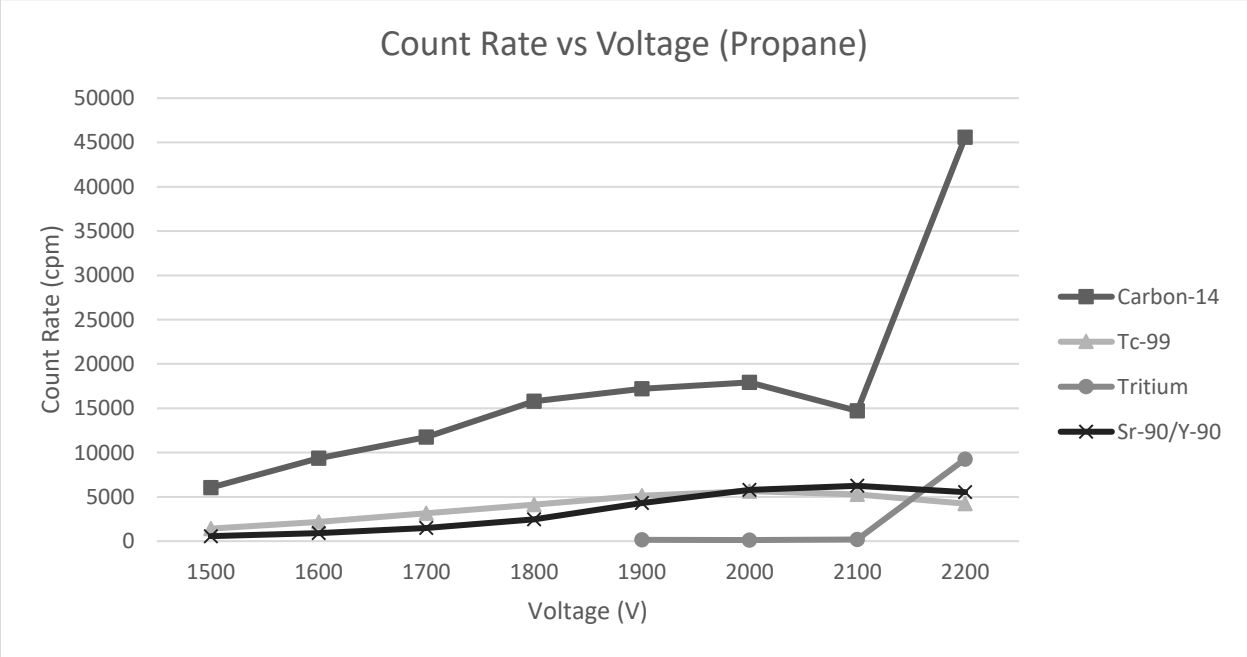

Figure 11. Count rate vs Voltage from propane gas tests.

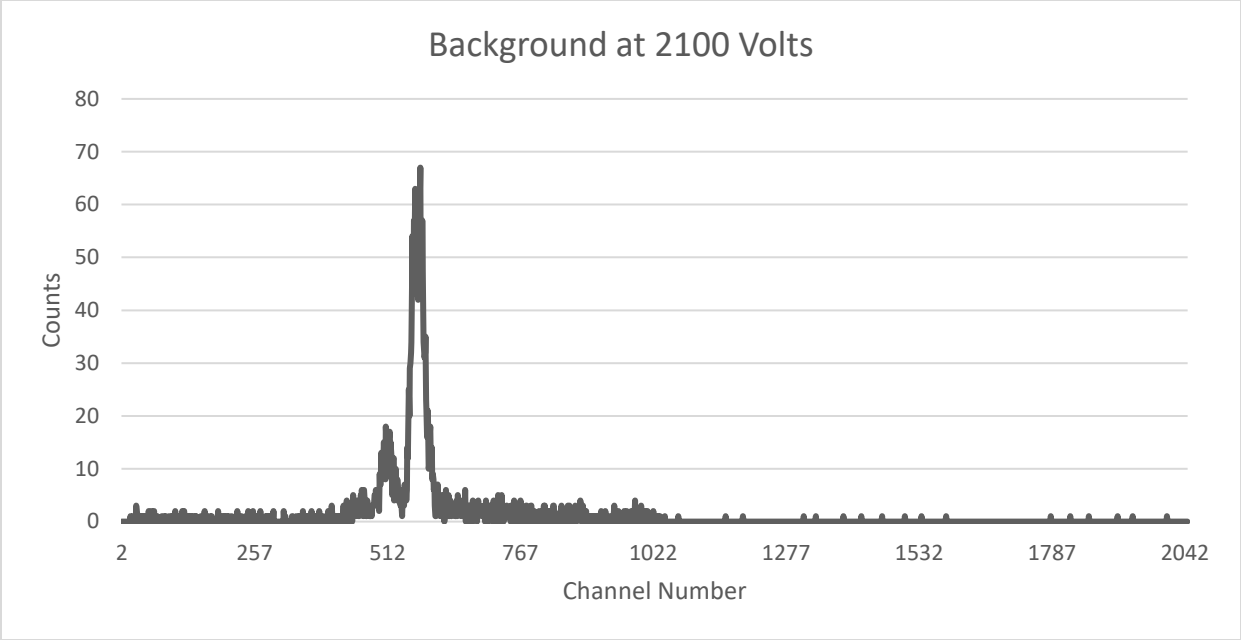

Figure 12. Overarcing in background reading for technetium-99 at 2100 volts.

Conclusion

The propane substitute gas in the constant flow proportional counter showed an increase in count rate as a function of increased voltage, providing evidence that gas multiplication is occurring. The spectra from propane was not as clear as the P10 gas because while there is gas multiplication, there is not enough from propane. There were not enough electrons being liberated in order to create large enough pulse heights for all beta ray energies to be shown. A possible solution for this could be to create a larger volume detector. This would allow more time for gas multiplication, allowing more electrons to reach the anode per beta ray, thus creating a larger pulse height. Another attempt at this experiment could be done using MAPP gas, another readily available and portable welding gas. The only source to not support this conclusion was tritium because the energy was too low to produce adequate count rates differentiable from electronic noise.

References Cited

Cember, H. and Johnson T. E., 2009, *Introduction to Health Physics,* 4th edition, McGraw-Hill.

Cross, W.G., Ing, H., Freedman, N., 1983, A short atlas of beta-ray spectra, *Phys. Med. Biol.,* Vol. 28, No. 11, p. 1251-1260.

Mantel, J., 1972, The Beta Ray Spectrum and the Average Beta Energy of Several Isotopes of Interest in Medicine and Biology, *International Journal of Applied Radiation and Isotopes,* Vol. 23, p. 407-413.

Tsoulfanidis, N., 1995, *Measurement and Detection of Radiation,* 2nd edition, Taylor & Francis, Washington D.C.