

Survey of Meteorite Samples for ^{92}Nb , ^{98}Tc , and ^{60}Fe Using Gamma Ray Spectroscopy

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1.0 Introduction

1.1 The Project

The focus of this senior project was the use of gamma ray spectroscopy to survey meteorite samples for ^{92}Nb , ^{98}Tc , and ^{60}Fe (six meteorite samples were surveyed during this project; a comprehensive list is attached as an appendix). The presence of measurable amounts of ^{26}Al (half-life 717,000 years) in meteorites leads astrophysicists to believe that ^{92}Nb , ^{98}Tc , and ^{60}Fe should also be present in detectable amounts, though they have not yet been found (note: at least one previous attempt has been made; in 1993, Aaron Ginn, a previous student of Dr. Grismore, performed a preliminary search for these isotopes using Dr. Grismore's multi-dimensional gamma ray spectrometer). Since the isotopes that were looked for in this senior project are not long lived in comparison to the age of the solar system, their presumed presence indicates that they are continuously being made in outer space and deposited on space objects. While the discovery of these isotopes by their characteristic gamma rays will provide empirical evidence to support current theory, a negative result—finding no evidence of these isotopes in any of the meteorite samples—is intriguing in itself; as with all physics, it will only stimulate further investigation.

1.2 Acknowledgements

The author would like to extend many thanks to his senior advisor Dr. Roger Grismore. Dr. Grismore's expertise and guidance allowed this senior project to be the excellent hands-on experience that Cal Poly is known for. It should also be noted that much gratitude is extended because Dr. Grismore made an effort to craft this project to fit the graduation time table of the author. The author would also like to thank Dr. Llewellyn and the University of Central Florida

for sending meteorite samples to be surveyed. Their cooperation allowed this project to be completed.

2.0 Background

2.1 The Meteorite Samples

It was desired to survey as many different types of meteorite samples as possible; hence both chondrites and stony-iron fragments were chosen. Four different types of meteorites were surveyed during this project: ordinary chondrite (Charsonville, Guareña, Lake Labyrinth), carbonaceous chondrite (NWA 2086), mesosiderite (NWA 2932), and coarse octahedrite (Canyon Diablo).

Chondritic meteorites are a stony type of meteorite that were formed by the accretion of dust and other particles in the early solar system. They are primarily composed of an aggregate of chondrules—individual grains formed by molten or partially molten droplets—rich in olivine and pyroxene and fine dust, some of which is made up of presolar grains. This type of meteorite contains the oldest material in the solar system and has thus been in Earth’s local vicinity for the longest period of time. These two characteristics make chondrites good candidates to study. The chondritic meteorites are also the most common and are easiest to acquire.

Ordinary chondrites are the most common of the chondrites and are classified into three mineralogically distinct groups: H, L, and LL. H-type ordinary chondrites are characterized by a high abundance of free iron. The Charsonville and Guareña meteorite fragments are H-type. L-type ordinary chondrites are so named for their relatively low concentration of free iron. LL-type ordinary chondrites are named for their low iron and low free-metal content. The Lake Labyrinth meteorite fragment is of type LL.

Carbonaceous chondrites are the most chemically pristine matter in the solar system; they are also very rare, making up less than five percent of witnessed meteorite falls. They formed in oxygen-rich environments in the solar nebula that eventually became the present day solar system. Unlike in ordinary chondrites, the metals in this type of meteorite are in the form of silicates, oxides, or sulfides. Some even contain water and organic compounds such as amino acids. The NWA 2086 meteorite fragment is a carbonaceous chondrite of type CV-3. The CV indicates that it is of the same type as the Vigarano meteorite that fell in Italy in 1910 and the 3 indicates the petrologic type. The fact that they have undergone little chemical or heat-induced changes since their creation is significant to a study like this senior project.

Mesosiderites, a class of stony-iron meteorites, are a breccia—made of broken fragments of minerals and rock held together by a fine-grained matrix—made of approximately equal amounts of silicates and nickel-iron. The silicates in mesosiderites are primarily olivine, pyroxene, and feldspar. The NWA 2932 meteorite sample is a mesosiderite and provides a good contrast to the chondrites.

Octahedrites are the most common of the iron meteorites and are primarily composed of iron-nickel alloys. Trace concentrations of gallium, germanium, and iridium are used to separate the iron meteorites into chemical classes. The Canyon Diablo fragment is a coarse octahedrite of type IAB—“coarse” referring to the size of iron-nickel crystals and IAB referring to the chemical classification—and provides a good contrast to the all the other stony samples.

2.2 The Radioisotopes

The subject unstable isotopes of iron, niobium, and technetium are extinct radionuclides—radionuclides that were present in the early solar system, but have since decayed away to undetectable levels. Most extinct radionuclides have short half-lives and are therefore

harder to detect; however, with half-lives of 34.7 Ma, 4.2 Ma, and 1.5 Ma respectively; ^{92}Nb , ^{98}Tc , and ^{60}Fe may be more easily detectable. Since the solar system is approximately 4.5 billion years old, these radionuclides, if found, are not primordial, but rather sourced from more recent supernova debris swept into the local interstellar medium or from cosmic ray spallation, a process of nucleosynthesis by which energetic cosmic rays bombard an atomic nucleus and blast off some of its nucleons, thereby producing a new element.

^{92}Nb decays almost entirely by electron capture following the scheme $^{92}_{41}\text{Nb} + e^- \rightarrow ^{92}_{40}\text{Zr} + \nu_e$, where the decay product $^{92}_{40}\text{Zr}$ is stable. In electron capture an atom “captures” one of its orbital electrons which leads to a proton being converted into a neutron with the emission of an electron neutrino. The resulting nucleus has the same mass number, but one less proton, which indicates a new element. The new element—in this case Zirconium—is in an excited state because one of the inner shell electrons is now missing and in the process of de-exciting to the ground state, emits multiple photons. For the decay of ^{92}Nb , these include gamma ray photons with energy 561.1 keV and 934.5 keV.

^{98}Tc undergoes beta minus decay following the scheme $^{98}_{43}\text{Tc} \rightarrow ^{98}_{44}\text{Ru} + e^- + \bar{\nu}_e$, where $^{98}_{44}\text{Ru}$ is stable. Beta minus decay refers to the spontaneous conversion of a neutron into a proton with the emission of an electron and an anti-electron neutrino. The excited $^{98}_{44}\text{Ru}$ then emits two gammas of energy 745.4 keV and 652.4 keV.

^{60}Fe undergoes beta minus decay following the scheme $^{60}_{26}\text{Fe} \rightarrow ^{60}_{27}\text{Co} + e^- + \bar{\nu}_e$. $^{60}_{27}\text{Co}$ then decays in the same way by $^{60}_{27}\text{Co} \rightarrow ^{60}_{28}\text{Ni} + e^- + \bar{\nu}_e$. Since the intensity of the gammas emitted following the decay of ^{60}Fe is low, the decay of iron’s decay product, ^{60}Co is focused upon. As the $^{60}_{28}\text{Ni}$ de-excites following the $^{60}_{27}\text{Co}$ decay, gamma rays (as well as x-rays) are

emitted, the most intense being those with energies 1173.2 keV and 1332.5 keV. This marks the end of the decay chain, as ${}^{60}_{28}\text{Ni}$ is stable.

2.3 The Spectrometer

The instrument used to detect characteristic gamma rays for this project is a high-purity germanium (HPGe) one-dimensional spectrometer surrounded by ten centimeters of lead shielding to block background radiation. The spectrometer has a semiconductor detector made of a single, highly pure crystal of germanium. Ionizing radiation, in this case gamma photons, enter the detector and induce electrons to move from the valence band to the conduction band thereby producing holes in the valence band. The number of electron-hole pairs is proportional to the energy of the incoming radiation. A high voltage source is connected to opposite ends of the detector to produce an electric field across it. This field causes the electrons and holes to be drawn to opposite electrodes where they produce a current signal that is then converted to a voltage and sent to the amplifier and then on to the computer.

Occasionally, the gamma photons will scatter off of the electrons in the detector. This results in only part of the original energy being imparted on the electron, with the rest being emitted in the form of a lower energy photon. This process is known as Compton scattering and produces a “Compton smear,” most noticeable in the lower energies of the measured spectrum.

Electron-positron pair production is another unintended effect that occurs in the detector. When photons with energies greater than 1.022 MeV (twice the rest mass energy of an electron) interact with an atomic nucleus in the detector, an electron-positron pair may be produced without violating the conservation of momentum. Within a very short amount of time, the positron will annihilate with an electron producing two gammas with energies of 511 keV. This effect is quite apparent when looking at the peaks in the spectrum.

One drawback to using the HPGe spectrometer is the need to keep the detector cooled to liquid nitrogen temperatures. This is necessary because the band gap energy in the germanium crystal is low enough such that thermal excitation of electrons at room temperature is enough to boost them into the conduction band, which results in debilitating amounts of noise.

3.0 Procedure

3.1 Set-up and Data Recording

The experimental set-up needed to conduct this project is rather simple and consists of the detector, cooled by a constant supply of liquid nitrogen, connected to a high voltage source and a signal amplifier. The signal amplifier is then connected to a personal computer analyzer (PCA) card (a type of multichannel analyzer) in the computer.

It is important to prevent any contamination when dealing with the samples whose spectra will be studied. This involved using latex gloves to prevent skin oils from being deposited on the samples and/or the plastic bags that they were contained in. Clean plastic zip-top snack bags were used to prevent any direct contact of the sample and/or its container with the surface of the spectrometer.

Before any runs are made, it is necessary to calibrate the instruments so that we can simply read the channel number in the spectrum as the energy of the photon that produced the signal. We do this by using a strong, mixed $^{137}\text{Cs}/^{60}\text{Co}$ source that produces peaks of known gamma emission energies (661.6 keV, 1173.2 keV, and 1332.5 keV) to adjust the gain on the amplifier and the baseline (zero adjustment) on the PCA card. The gain is adjusted so that each channel represents 1 keV and the baseline is adjusted so that 0 keV is represented by channel zero. 2048 channels were used for this project as all the peaks of the isotopes searched for are less than 2048 keV.

After calibration, the experimental runs were made over the course of a few months. The first of these was a background run with just a zip-top plastic snack bag. All but one of the runs was about two weeks long (exact times are found in the appendix). Each time a new sample was surveyed, after a couple days of data were taken, a feature in the PCA software called the digital spectrum stabilizer (DSS) was activated to digitally correct for gain and baseline (zero) drift. This stabilizes the position of the peaks and allows for confidence that the centroid of the peaks have not changed over the course of the run. Once the raw spectra were taken, analysis using software written by Dr. Grismore allowed the author to look for peaks corresponding to the characteristic gammas of ^{92}Nb , ^{98}Tc , and ^{60}Fe .

3.2 Interpreting the Raw Data

Once the runs were completed, the spectra were read off as ASCII files and processed using Microsoft Excel to create a file with just the count data in 2048 rows corresponding to the 2048 channels used. QUADTRFM.EXE is then used to obtain the three coefficients necessary to adjust the spectrum. This program is a quadratic extension of the program described in the article "Internal Energy Standardization of Multichannel Gamma Ray Spectra by Use of the 0.511 MeV Annihilation and K-40 Peaks" by R. Grismore, et al. It takes the positions of three peaks of known energies 186.05 keV, 911.205 keV, and 1764.494 keV associated with ^{226}Ra , ^{228}Ac , and ^{214}Bi , respectively, and fits them in quadratic fashion to the positions of those peaks in the sample spectrum. 2ADJ2048.EXE is then used, along with the three coefficients from the output of QUADTRFM.EXE, to shift the sample spectrum to produce a calibrated spectrum from which we can subtract the background. SUBTRACT.EXE is used to produce a final set of numbers which are graphed versus channel number. The output of this program is sample spectrum minus the background. This data set is then examined for the peaks associated with the characteristic

gammas from ^{92}Nb , ^{98}Tc , and ^{60}Fe or any other interesting peaks. If any peaks are found, we apply the standard count data uncertainty for N counts, which is $\pm\sqrt{N}$.

4.0 Results

It should be noted that there are some commonly observed gamma peaks that are likely to be seen in most spectra as shown in the following table.

Energy (keV)	Isotope
1764	Bi-214
1461	K-40
1120	Bi-214
969	Ac-228
911	Ac-228
609	Bi-214
583	Ti-208
511	e^-e^+
352	Pb-214
295	Pb-214
241	Ra-226

Table 1. Commonly observed gamma energies seen in sample spectra.

Out of the six meteorite samples that were surveyed, peaks corresponding to the characteristic gammas from ^{92}Nb , ^{98}Tc , and ^{60}Fe were observed in none of the spectra, a disappointing, but not completely unexpected outcome. However, it is interesting to note that a peak near 1808 keV was observed in the spectra from the Guareña and Charsonville fragments. This peak is indicative of ^{26}Al (as it decays to ^{26}Mg), the subject of study for previous students of Dr. Grismore.

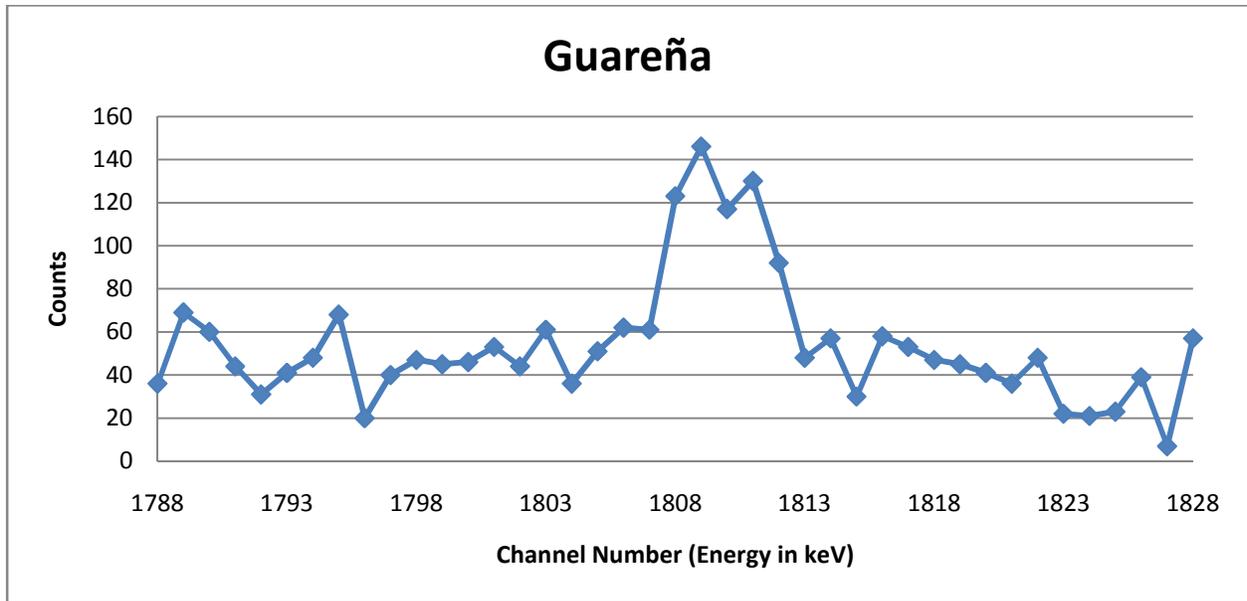


Figure 1. ^{26}Al peak in the spectrum from the Guareña fragment.

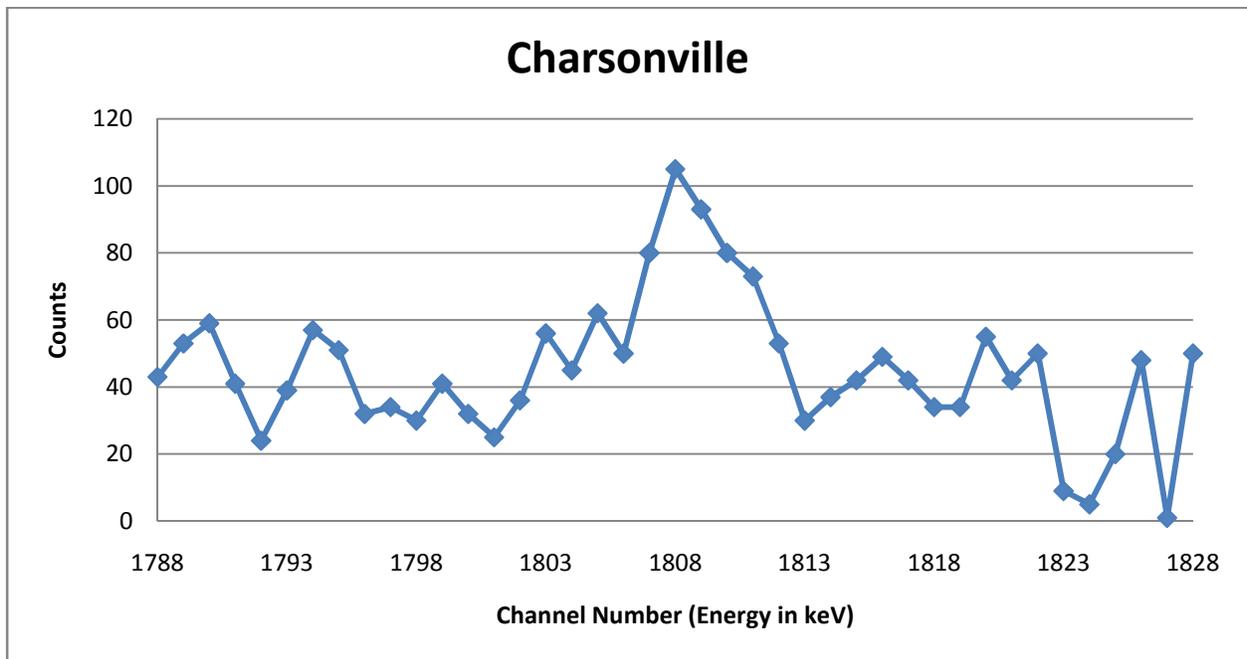


Figure 2. ^{26}Al peak in the spectrum from the Charsonville fragment.

Also seen was a small peak near 1181 keV in the Charsonville spectrum. This was interesting because all known gammas with a range of 1179 to 1183 keV come from short lived

isotopes (half-lives < 1 year), meaning that either this was an improbable spike in the background that was not a real peak at all or that some sort of contaminant had been detected.

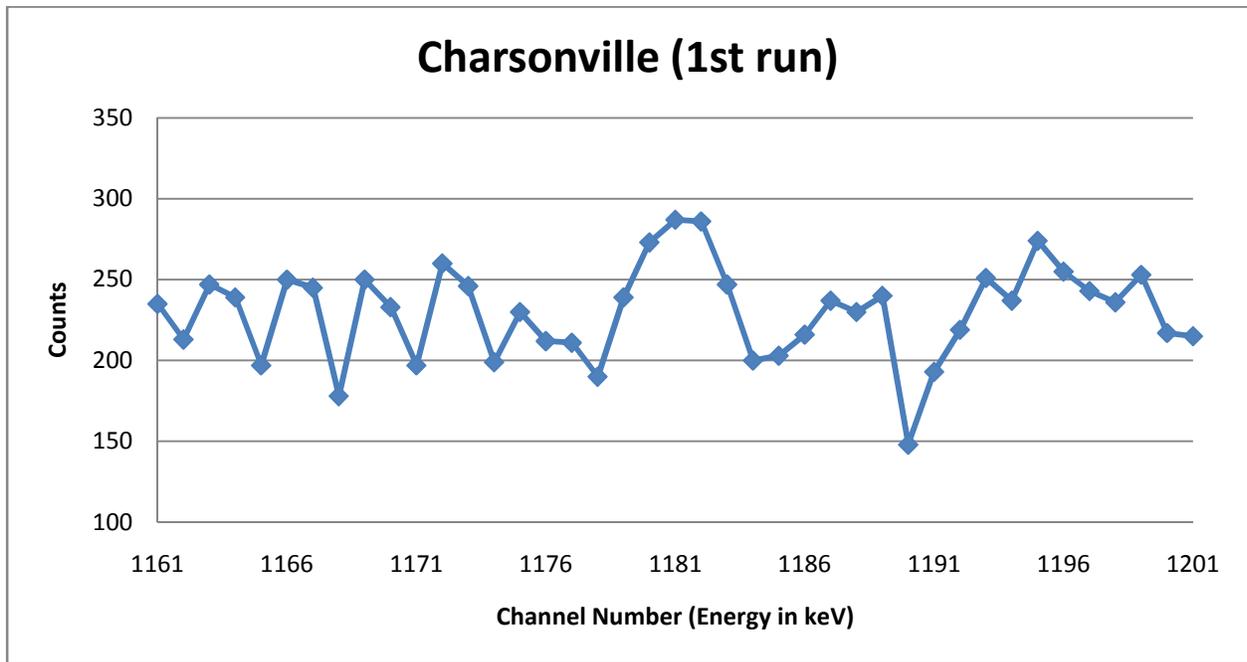


Figure 3. Unknown peak in the Charsonville spectrum.

In the interest of possibly finding out more information, a second run was made to see if any decay had occurred.

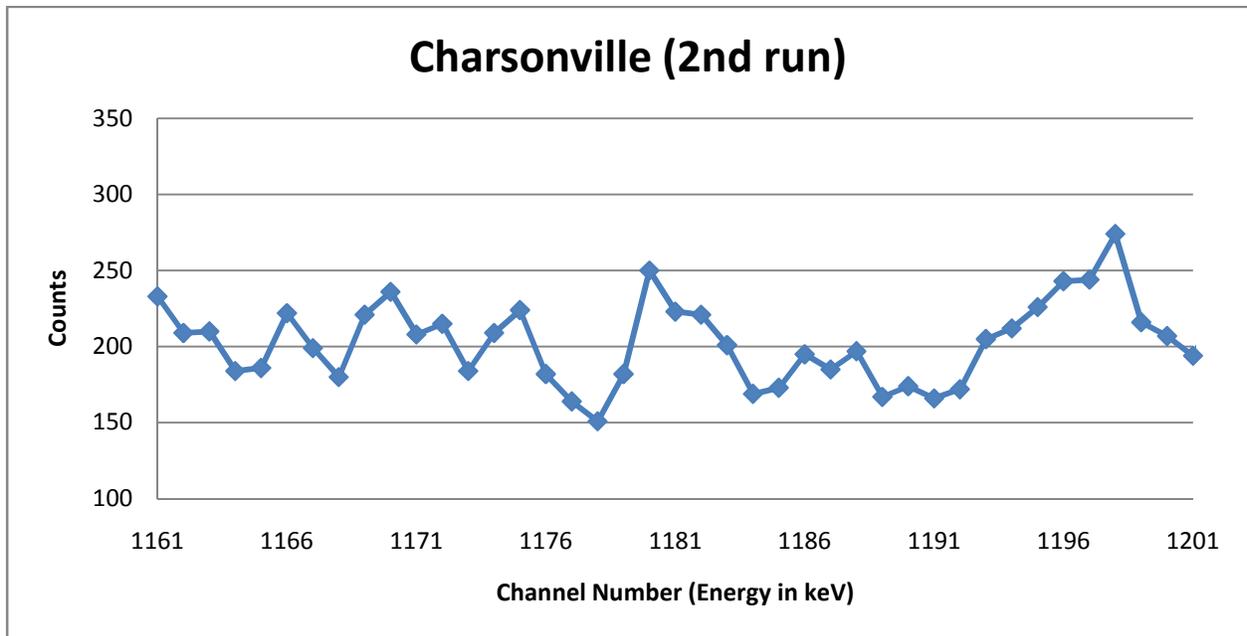


Figure 4. The same area in the Charsonville spectrum after a second run.

If we take the average number of counts per channel within twenty channels of the proposed peak at 1181 keV, then take the difference between that number and the number of counts in channels 1179 to 1183, we can find the total number of counts that comprise the peak. Since the run times are slightly different, the number of counts for the second run must be multiplied by the ratio of the first run time to the second run time. For the first run, this comes to 112 counts and for the second run the number is 41 counts. We can now use the familiar exponential decay equation to find the experimental half life with the knowledge of the time between the first and second runs.

$$N = N_0 e^{-\lambda t} \text{ and } N_{t_{1/2}} = N_0 e^{-\lambda t_{1/2}}$$

Solving for λ in each equation, equating them, and then solving for $t_{1/2}$ gives an equation for the experimental half life:

$$t_{1/2} = \frac{\ln(2) t}{\ln\left(\frac{N_0}{N}\right)}$$

where t is the time between runs (3,196,000 seconds), N_0 is the number of counts in the peak in the first run (112) and N is the number of counts in the peak in the second run (41). Factoring in the uncertainty of the counts and performing error propagation, this gives an experimental half life of 25.5 ± 4.6 days. The closest isotope match is ^{147}Eu , which has a gamma energy at 1180.231 keV and a half life of 24.1 days.

5.0 Conclusions

Although no evidence of ^{92}Nb , ^{98}Tc , and ^{60}Fe was found in this investigation, it was still worthwhile to start looking and a confirmation of the presence of ^{26}Al was encouraging. Perhaps with a more sensitive spectrometer and/or lengthier run times the peaks will be noticeable. In

hindsight it would probably have been a better idea to survey meteorite fragments with larger masses and surface areas to maximize the amount of material surveyed as well as the amount of meteoric surface exposed to particles in space that might harbor the isotopes that were looked for.

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Appendix

Meteorite Fragment Information			Run Times (s)		QUADTRFM.EXE Output		
Sample	Mass (g)	Description	Live Time	Real Time	A	B	C
Lake Labyrinth	20.2	Ordinary chondrite. Fell, May 2, 1924, Lake Labyrinth, South Australia. Grey stone fragment, brown inside core, brown flecks on the exterior.	1947520	1947673	-2.01044E-07	1.0008204	-1.3156749
Guareña	167.0	Ordinary chondrite. Fell, July 20, 1892, Badajos Spain. Irregular fragment with partial fusion crust.	1370490	1370581	-4.15879E-07	1.00050461	-1.3394866
Charsonville	89.0	Ordinary chondrite. Fell, November 23, 1810, Meung, Loiret, France. Irregular fragment.	1st run: 1270134 2nd run: 1200698	1st run: 1270190 2nd run: 1200749	1st run: -5.078923E-06 2nd run: -3.06083E-08	1st run: 0.99977815 2nd run: 0.99954408	1st run: -0.85113835 2nd run: -0.71410447
NWA 2086	26.1	Charbonaceous chondrite (CV3). Found November 2003 in the Sahara desert of North Africa. Similar to Allende with early nebular condensates and interstellar material that gives clues to the solar system's formation.	600068	600094	-7.06818E-07	1.0006032	-1.13776278
NWA 2932	50.7	Mesosiderite. Found in the Sahara desert of North Africa. This meteorite consists of a collection of silicate rock fragments set in a matrix of metal. It was probably formed during an impact.	1208395	1208446	-4.11563E-07	1.0008446	-1.7028842
Canyon Diablo	500.0	Iron, Coarse Octahedrite (IAB). Fragment a remnant of a ~40 meter diameter asteroid that hit Arizona over 50,000 years ago.	1075592	1075672	-9.58469E-07	1.00165164	-1.95410633