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**Determination of Silver Using
Cyclic Epithermal Neutron Activation Analysis**

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Supervisor:

Sheldon Landsberger

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by

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Thesis

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Abstract

Determination of Silver Using Cyclic Epithermal Neutron Activation Analysis

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The University of Texas at Austin, 2010

Supervisor: Sheldon Landsberger

A fast pneumatic transfer facility was installed at the Nuclear Engineering Teaching Laboratory (NETL) of the University of Texas at Austin for the purpose of cyclic thermal and epithermal neutron activation analysis. In this work efforts were focused on the evaluation of cyclic epithermal neutron activation analysis (CENAA). Various NIST and CANMET certified materials were analyzed by the system. Experiment results showed ^{110}Ag as one of the isotopes favored by the system. Thus, the system was put into practical application for identifying silver concentration in the Arctic atmospheres in air filters collected in 2009, and traces in metallic ores. Comparison of silver concentrations via CENAA with the CANMET certified reference materials gave very good results.

Table of Contents

Acknowledgements.....	iv
Abstract.....	v
Table of Contents.....	vi
List of Tables	viii
List of figures.....	ix
1 Introduction.....	1
1.1 Neutron Activation Analysis.....	1
1.2 Advantages and Limitations	2
1.3 Common NAA Applications.....	4
1.4 Development of Neutron Activation Analysis with Respect to Cyclic Systems	6
1.5 Tracking of Silver Using Cyclic Epithermal Neutron Activation Analysis	9
2 Theory.....	10
2.1 Neutron Activation Analysis.....	10
2.2 Cyclic Neutron Activation Analysis	12
2.3 Epithermal Neutron Activation Analysis.....	15
3 Experimental.....	20
3.1 Experimental Design.....	20
3.2 Experiment Apparatus	21
3.3 Sample Preparation	30
3.4 Setup of the Pneumatic Transfer System	33
4 Results and Discussion	37
4.1 Neutron Flux Statistics.....	37
4.2 Biological and Coal Samples	40
4.3 Metallic Ore Specimens.....	47
4.5 silver standard reference and concentration calculation	58
4.6 Source of Uncertainty and Error	62
5 Recommendations and Future Work	64
5.1 Dead-Time Correction	64

5.2 Sample Transfer Time.....	64
6. Conclusions.....	66
Bibliography	67
Vita	69

LIST OF TABLES

Table 1. Sensitivities of Detection with a 10^8 n / cm ² Thermal Neutron Flux	3
Table 2. Element Contents in Certified Reference Materials	5
Table 3. NAA Result of Silver Coins	6
Table 4. Cadmium Ratio of Common Elements	9
Table 5. Sample Contents and Purposes	32
Table 6. Elements in Biological Samples	45
Table 7. Elements in Metallic Ore Specimens	51
Table 8. Signal-to-Background Ratio for One and Two NAA Cycles of Zn-Pb-Sn-Ag Ore	52
Table 9. Net Area Counts of Standard Reference Solution	58
Table 10. Silver Concentration in Ores Specimens	61
Table 11. Photon Attenuation Percentage in Different Ore Specimens	63

LIST OF FIGURES

Figure 1. Neutron Total Cross Section of ^{27}Al	2
Figure 2. Neutron total cross-section of ^{113}Cd	8
Figure 3. Signal-to-Background Ratio of Conventional and Cyclic NAA	15
Figure 4. Energy Distribution of Prompt Fission Neutrons	16
Figure 5. Neutron Energy Distribution of Neutrons Reaching the Irradiation Chamber.....	18
Figure 6. Neutron Total Cross-Section of ^{109}Ag	19
Figure 7. Loading Dock Sectional Diagram	22
Figure 8. Loading Dock	23
Figure 9. Functional Diagram of the Automated Pneumatic System	24
Figure 10. Functional Diagram of the Automated Pneumatic System	25
Figure 11. An Overview of the Entire System.....	25
Figure 12. PLC Com User Panel.....	28
Figure 13. Polyethylene Vials.....	29
Figure 14. Gamma Spectrum of an Aluminum Wire.....	38
Figure 15. Fluctuation of Neutron Flux	39
Figure 16. Gamma Spectrum of Tomato Leaves	41
Figure 17. Gamma Spectrum of Apple Leaves.....	41
Figure 18. Gamma Spectrum of Peach Leaves	42
Figure 19. Gamma Spectrum of Pine Needles.....	42
Figure 20. Gamma Spectrum of Rice Flour.....	43
Figure 21. Gamma Spectrum of Wheat Flour.....	43
Figure 22. Gamma Spectrum of Bovine Liver.....	44
Figure 23. Gamma Spectrum of Coal Trace Element.....	46
Figure 24. Gamma Spectrum of Gold Ores	47
Figure 25. Gamma Spectrum of Nickel-Copper Ore	48
Figure 26. Gamma Spectrum of Nickel-Copper-Cobalt Ore 1	48
Figure 27. Gamma Spectrum of Zinc-Lead-Tin-Silver Ore	49
Figure 28. Gamma Spectrum of Zinc-Tin-Copper-Lead Ore	49
Figure 29. Gamma Spectrum of Concentrated Lead Ore	50
Figure 30. Gamma Spectrum of Concentrated Zinc Ore	50

Figure 31. Gamma Spectrum of Arctic Air-Filter (Taken on January 7, 2009)	53
Figure 32. Gamma Spectrum of Arctic Air-Filter (Taken on July 7, 2009)	54
Figure 33. Backward Trajectory Model of Arctic Air Flow	55
Figure 34. Airborne Silver Concentration in the Year of 2009	56
Figure 35. Silver and Aluminum Trend over Summer of 2009	57

1 Introduction

1.1 NEUTRON ACTIVATION ANALYSIS

Neutron activation analysis (NAA) is a nuclear analytical method used to determine elemental concentrations within a mixture of various substances. NAA was first discovered in 1936 by George de Hevesy and Hilde Levi using neutrons from a radium-beryllium source (Frame 1997). Starting from 1950s, through with the advancement of nuclear technology and increase in the number of nuclear research reactors, NAA had been employed world-wide for a broad diversity of specimens.

NAA makes use of a common neutron interaction with matter called radiative capture. During this interaction, the target nuclides absorb neutrons and become unstable. The unstable nuclides undergo beta decay along with a gamma ray emission from the ground state. The majority of NAA experiments are practiced with thermal neutrons, which have an average energy of about 0.025 eV with a corresponding speed of 2200 m s^{-1} (Shultis and Faw 2000). Thermal neutrons are used for two reasons. The first reason is that thermal neutrons are the most available through fission as they are thermalized. NAA is often carried out at research nuclear reactors, where uranium fuel is allowed to fission to generate neutrons. For research reactors, more than 90% of neutrons in the moderated neutron flux possess energy level below 1 eV (Shultis and Faw 2000). The other reason for using thermal neutrons is that many nuclides are found with higher thermal absorption cross-sections. This is due to the fact that neutron cross-sections of most nuclides have a one-over-velocity correlation. The correlation is illustrated in Figure 1, a plot of ^{27}Al neutron cross section versus neutron energy. As can be seen the cross section is high at low neutron energy. Usually resonance peaks at high neutron energies have negligible importance to NAA since there are less likely to be produced. However, many nuclides do exhibit resonances in the epithermal range.

Figure 1. Neutron Total Cross Section of ^{27}Al (NNDC 2006)

1.2 ADVANTAGES AND LIMITATIONS

NAA has been defined as one of the best multi-elemental analytical methods.

- (1) NAA is capable of determining multiple elements in a single analysis. NAA has been proven effective on stable isotopes from more than 70 elements (Kruger 1971).
- (2) NAA has a high sensitivity and is capable to determine elemental concentrations from a few nanograms to micro-grams to milligrams of sample mixture (Glascock 2010). A number of common NAA target nuclides are listed in Table 1.

Table 1. Sensitivities of Detection with a 10^8 n / cm² Thermal Neutron Flux (Taylor 1964)

Limit of Detection (ppm)	Elements
0.1 to 1	V, Co, Br, Au, In, I, Mn
1 to 10	Na, Al, Cl, Cu, As, Sb, W
10 to 50	Ti, Mo, Ru, Pt
50 to 100	Mg
100 to 400	K, Ca, Ni, Zn

(3) NAA is non-destructive to samples. Despite the fact that activated nuclides undergo beta decays and become another radionuclide, the influence to physical and chemical properties of the sample is negligible. This is because only an extremely small portion of the sample nuclides are activated during irradiation. Therefore, samples analyzed by NAA may be reused in other analytical techniques.

(4) NAA is instrumental ready. Both solids and liquids are set to be irradiated and counted afterwards. There is no chemical preparation required before the analysis. This saves time, resources and while limiting the potential opportunities that samples get contaminated. Sample preparation procedures will be further discussed in Chapter 4.

(5) Multiple samples can be analyzed by employing automated instrumentation. The simple mechanism of NAA allows the process to be performed and controlled by computer programs. This allows NAA to be carried out in large scales with minimum labor and time.

On the other hand, there are several limitations to NAA that should be remarked.

(1) NAA requires a neutron source such as nuclear reactor, which is not commonly available in many parts of the world. Nuclear reactors also require a high maintenance cost and the need to purchase uranium fuel.

(2) NAA is not as effective for nuclides with low absorption cross section and to those that have a low gamma-ray activity emission.

(3) Samples that are analyzed by NAA are radioactive and are to be set aside to “cool off” before the samples are safe to be handled for other experiments or need to be permanently disposed.

1.3 COMMON NAA APPLICATIONS

The purpose of NAA is to quantify the amount of an element within a specimen. As discussed in section 1.2 above, NAA has a high sensitivity and is capable of determining many elements. Therefore, this technique can be used to analyze geological, environmental, biological and material samples for various researches. One example is the determination of elements in different food samples. During the 1970s, there was increasing interest in food composition for nutrition, along with amount of toxic substances such as heavy metals. One common element to be tracked within food is selenium, which is an essential nutritional element (Chatt, et al. 1988). Lack of selenium may cause various liver, kidney and heart diseases. Therefore, many nutritionists would desire to know the amount of selenium in different diets. Chatt (1988) had performed cyclic instrumental NAA for hospital diets to track selenium. He also compared his NAA results to the reported values from reference materials as seen in Table 2. The precision of the measurements was within 5%-7% for most of the specimens. It was also concluded that the selenium detection limit was as low as 3 ppb (parts per billion) for simple food ingredients, whereas it was 29 ppb for samples with high salt concentration (Chatt, et al. 1988). Besides selenium, food samples are often studied for iodine, chlorine, sodium and aluminum.

Table 2. Element Contents in Certified Reference Materials (Chatt, et al. 1988)

Elemental content of mixed diets by INAA				
Element, unit	Mixed Human Diet (IAEA H-9)		Mixed Diet (NBS RM-8431)	
	This Work*	IAEA Values	This Work	Recomm. Values
Al, ppm	10 ± 2	----	4 ± 1	4.39 ± 1.07
As, ppb	----	88 ± 33	1000 ± 320	924 ± 344
Br, ppm	7.72 ± 0.75	7.5 ± 0.7	9.79 ± 0.41	----
Ca, %	0.225 ± 0.021	0.231 ± 0.016	0.180 ± 0.012	0.194 ± 0.014
Cl, %	1.45 ± 0.11	1.25 ± 0.15	0.558 ± 0.006	----
Co, ppb	48.5 ± 5.3	43 ± 5	36.3 ± 2.4	37.6 ± 7.8
Cr, ppb	115 ± 20	150 ± 40	103 ± 11	102 ± 6
Fe, ppm	33.7 ± 2.5	33.5 ± 2.5	35.8 ± 3.4	37.0 ± 2.6
K, %	0.820 ± 0.065	0.830 ± 0.07	0.789 ± 0.10	0.790 ± 0.104
Mg, %	0.078 ± 0.007	0.0785 ± 0.0055	0.066 ± 0.003	0.065 ± 0.004
Mn, ppm	11.3 ± 1.2	11.8 ± 0.8	7.83 ± 0.40	8.12 ± 0.31
Na, %	0.813 ± 0.075	0.81 ± 0.07	0.320 ± 0.006	0.312 ± 0.016
Rb, ppm	7.65 ± 0.80	8.0 ± 0.6	6.65 ± 0.42	----
Sb, ppb	2.5 ± 0.4	----	7.4 ± 2.4	----
Sc, ppb	0.62 ± 0.20	----	0.67 ± 0.23	----
Sn, ppm	----	----	58.4 ± 2.5	----
Zn, ppm	28.2 ± 0.4	27.5 ± 1.8	18.1 ± 0.9	17.0 ± 0.6

Studies had also suggested that appetite control may be influenced by metal content, such as zinc, manganese and copper, consumed along with intake of food. NAA was used correlate these elements with obesity (Spyrou 1999)

Besides nutrition, NAA has also been applied in the biomedical field to track diseases and syndromes in body. NAA helps indentifying an unbalance of life essential elements and over dose of toxic substances. For instance, patients with Alzheimer's disease are suspected of having elevated aluminum in their brains, kidneys and livers (Spyrou 1999)

NAA is a powerful tool in the category of scientific crime investigation. NAA is employed to determine various elements from items left at the crime scene. Even the smallest fragment of an evidence may help lead to identify the criminal responsible for the crime. For instance, tracking the metal content of a bullet or bullet shell identifies the category of arms. Furthermore, prompt gamma NAA techniques are considered for explosive detection due to its

ability to determine nitrogen. Such techniques can also be applied to identify landmines laid over battlefields.

Another common application is the determination of rare or commercial metals. Soil or rock samples from potential mining sites are analyzed for copper, iron, tin, gold, silver, platinum or even uranium. Besides mining purposes, metal determination is proven useful in many other ways. NAA was used to analyze ancient silver coins in order to determine the weight of silver and copper in each batch of coins (Elayi, Damiani and Collet 1985). From their findings in the report, as shown in Table 3, archaeologists were able to track the origin and make of the coins.

Table 3. NAA Result of Silver Coins (Elayi, Damiani and Collet 1985)

Class number of the coins, die numbers and the results of the analysis for copper and silver					
Class number	Obverse die number	Coin number in the hoard	Reverses linkage	Weight of silver, g	Weight of copper, g
III	D7	8		2.54	3.59
III	D4	11		2.60	3.36
III	D17	46		2.28	3.74
III	D17	47	}	2.40	3.30
III	D17	49		2.31	3.75
III	D17	48	}	2.25	3.45
III	D17	50		2.32	3.53
III	D13	34		2.11	3.60

1.4 DEVELOPMENT OF NEUTRON ACTIVATION ANALYSIS WITH RESPECT TO CYCLIC SYSTEMS

In 1966, Caldwell suggested cyclic neutron activation analysis (CNAA) as a combination neutron experiment for lunar and planetary samples (Spyrou and Kerr 1979). Samples are irradiated and counted in consecutively repeated cycles under the same conditions. The total response from all cycles is obtained by summing up the count measurements from each cycle. The purpose of the repeated cycles is to improve the detection limits and signal-to-background ratio of short-lived isotopes. CNAA is often carried out under an automated instrument in order to keep track of a precise combination of irradiation time, decay time and count time in each of

the repeated cycles. Therefore, it is sometimes referred as instrumental cyclic neutron activation analysis. CNAA has been used with different neutron sources such as neutron generators, isotopic sources and pulsed accelerator neutron beams for various application purposes in field of biochemistry and geology.

Spyrou was one of the original pioneers in cyclic activation analysis. In one of his papers, he discussed the concept of CNAA reducing the effect of background radiation (Spyrou and Kerr 1979). CNAA has an increasing peak to background ratio while conventional NAA has a decreasing ratio along with experimental time, due to the fading of radiation emitted by short-lived nuclides. Another way to suppress the signal from long lived background is to reduce the decay time between irradiation and counting. This is due to the fact that short-lived nuclides decay faster at a higher emission rate than long-lived ones.

NAA is also performed using epithermal neutron, with energies higher than 1 eV. As discussed in Section 1.1, typically neutron absorption cross section decreases as the energy increases. To perform NAA with epithermal neutrons, a cadmium tube can be inserted between the core and the irradiation chamber to remove thermal neutrons from the source. ^{113}Cd , which is one of the stable isotopes in cadmium, is a very effective neutron removal due to its unique cross section to thermal neutrons Figure 3 shows an ^{113}Cd total cross-section spectrum. ^{113}Cd has a very high thermal cross-section of above 20000 barns for thermal neutrons with energy below 0.5 eV. However, the cross-section drops dramatically to below 1000 barns above 0.5 eV. This 0.5 eV cutoff is often called the “cadmium cutoff”. In other words, neutrons with energy less than 0.5 eV are much more likely to be absorbed by the cadmium and never reach the sample in the irradiation chamber; while neutrons with high energy get to pass through.

Figure 2. Neutron total cross-section of ^{113}Cd (NNDC 2006)

To illustrate the effect of epithermal neutrons, Spyrou *et al.* (1982) compared the activity of twelve elements from the elemental standards (Na, Al, Sc, Ge, Se, Rh, Pd, Ag, Ce, Er, Hf, Pt) from a neutron beam through a bare tube and a cadmium sleeved tube respectively. Table 2 illustrates the result in terms of cadmium ratio, which is the ratio of activity of the isotopes in the bare tube to that in the cadmium tube.

$$R_C = \frac{\text{Activity due to irradiation in bare tube}}{\text{Activity due to identical irradiation in cadmium sleeved tube of the same specimen}}$$

The cadmium ratio indicates the magnitude of activity suppression by using cadmium to absorb thermal neutrons out of the irradiation beam. A high ratio would mean the activity of the isotope relies heavily on thermal neutrons and by shielding off thermal neutrons the activity of the isotope is reduced dramatically. On the other hand, a low ratio indicates shielding off thermal neutrons has little effect on the activity of the isotope. Therefore, ENAA is most effective when the isotope in interest has a low cadmium ratio while other isotopes within the sample have high ratio values as seen in Table 4.

Table 4. Cadmium Ratio of Common Elements

Isotope	Gamma energy	Half-life ¹	Cadmium ratio ²
²⁴ Na	1368.4	14.69 hour	18.5
²⁸ Al	1778.9	2.24 min	48.5
^{46m} Sc	142.5	18.75 sec	29.4
^{75m} Ge	139.8	47.7 sec	8.28
^{77m} Ge	215.5	52.9 sec	3.4
^{77m} Se	161.9	17.36 sec	33.9
¹⁰⁴ Rh	555.8	42.3 sec	4.7
^{104m} Rh	97.2	4.34 min	4.28
^{109m} Pd	188.9	4.7 min	1.02
¹⁰⁸ Ag	632.9	2.37 min	7.9
¹¹⁰ Ag	657.8	24.6 sec	2.39
^{139m} Ce	757	54.8 sec	3.9
^{167m} Er	208	2.27 sec	4.92
^{178m} Hf	428	4 sec	2.4
^{180m} Hf	215	5.5 hour	2.71
¹⁹⁹ Pt	319	30.8 min	3.1

¹ (KAERI 2000)² (Spyrou, et al. 1982)

1.5 TRACKING OF SILVER USING CYCLIC EPITHERMAL NEUTRON ACTIVATION ANALYSIS

Spyrou *et al.* (1982) stated and discussed the possibility of a cyclic epithermal neutron activation analysis (CENAA) with an automated pneumatic system controlled by computers. By using epithermal over thermal neutrons, signals from the long-lived nuclides are effectively suppressed while the signal ratio of short-lived is enhanced CENAA is therefore combining cyclic NAA and epithermal NAA. Despite a history of almost 30 years, published data using CENAA is rare.

2 Theory

2.1 NEUTRON ACTIVATION ANALYSIS

In general, NAA involves two steps: irradiation and counting. During NAA, the sample is first irradiated by a stream of neutrons. During the stage of irradiation, neutrons are allowed to interact with the sample nuclei. There are two main types of interactions between neutron and a nucleus: scattering and capture.

Scattering occurs when a neutron strikes a nucleus and leaves. A scattering is defined as “elastic” if the struck nucleus was left unchanged in both isotropic composition and internal energy. Elastic scattering is denoted as (n, n), indicating the incident neutron and the scattered neutron. On the other hand, a scattering is defined as “inelastic” when the struck nucleus was left in an excited state without a change in composition. The excited nucleus often decays by a gamma emission. Inelastic scattering interaction is denoted as (n, n') (Lamarsh 2002).

Neutron capture takes place as a neutron strikes and sticks to the nucleus of an atom. Due to the extra mass and kinetic energy of the incident neutron, the target nucleus becomes unstable and seeks to achieve stability through decays. Depending on the species of the atom, nucleus may decay in through various pathways. For NAA, the pathway in most interest is the gamma decay, in which the excited nuclide de-excites with the emission of a gamma ray. Such neutron capture and gamma emission is defined as radiative capture and is denoted by (n, γ). During the interaction, the target nucleus ^A_Z , with initial atomic number A, captures an incident neutron to form $^{A+1}_Z^*$. The '*' symbol here indicates the nucleus is in an excited state and is unstable. The nucleus emits the excited energy in form of a gamma particle and becomes $^{A+1}_Z$. Radiative capture can be expressed as Equation 1.

The energy level of the gamma emitted by the nucleus varies to different isotopes. This can be explained with a fundamental physics law: the conservation of energy. Equation 1 can be simplified into

For the purpose of reference, equation 2 is expressed as

where a and b are the reactants with c and d as products. The total energy before the reaction is sum the rest-mass energy and kinetic energy of a and b . Rest-mass energy of a particle is donated by the mass-energy equivalence formula, $E = mc^2$, where m is the rest mass and c is the speed of light. With similar approach, the total energy of the reaction product is the sum of energy of c and d . The energy equation becomes

$$m_a c^2 + m_b c^2 + E_a + E_b = m_c c^2 + m_d c^2 + E_c + E_d \quad (4)$$

Gamma particles are photons and have no rest mass (despite some recent studies show evidence photon may have a tiny mass, which is negligible in this case). Therefore, m_d equals to zero and E_d equals to the energy level of the gamma radiation. To find the energy level of the gamma emission, equation 4 can be rearranged as

$$E_d = (m_a + m_b - m_c) c^2 + E_a + E_b - E_c \quad (5)$$

However, this gamma radiation is often not accounted in NAA since it exists immediately after the interaction and only lasts for a small time frame. Once the sample leaves the irradiation chamber, these gamma activities will have already died out before the sample reaches the detector for counting.

Instead, NAA relies on delayed neutron emission for elemental detection. Many of the ${}^{A+1}Z$ nuclides formed during radioactive capture is still unstable in their composition after the emission of their excess energy. To achieve a stable state, the nuclides often undergo beta minus decay.

Equation 6 shows the reaction equation and product particles of the beta decay. A ${}_{Z-1}^AZ^*$ nuclide is formed with an emission of electron and antineutrino. Similar to the excited nuclide in radioactive capture, the excess energy is then emitted in form of gamma radiation, given by equation 7.

In NAA almost all isotopes with one or more gamma rays. Therefore, it is possible to identify each isotope by simply measuring the energy of a gamma ray emission with the proper instruments.

The method described above relies on delayed gamma radiation from radioactive capture. Methods designed for prompt gamma rays are not discussed in this thesis as it involves different concepts.

2.2 CYCLIC NEUTRON ACTIVATION ANALYSIS

CNAA is an integrated technique of consecutive irradiation-decay-and-count cycles. The measurement of CNAA is the cumulative radiation response of all the consecutive cycles. Each cycle can be divided into four periods: irradiation, transferring from reactor core to detector, counting, returning to core (Spyrou, *et al.* 1979). Therefore, each cycle period has four compositions as shown in equation 8.

$$T = t_i + t_t + t_c + t_r \quad (8)$$

where T - cycle period,

t_i - irradiation period,

t_t - transfer period, between end of irradiation and start of counting,

t_c - counting period,

t_r - return period, between end of counting and start of next irradiation

Assuming a specimen with a number of target nuclei N , decay constant λ and cross-section σ , is irradiated by an isotropic neutron flux Φ , the number of activated nuclei after the irradiation period can be expressed as

$$N_A = N\sigma\Phi (1 - e^{-\lambda t_i})$$

After the transfer period, number of activated nuclei left is

$$N_A = N\sigma\Phi (1 - e^{-\lambda t_i})(e^{-\lambda t_t})$$

Response about the radiation of interest with intensity I , counted by a detector with efficiency ϵ is therefore

$$D = \frac{N\sigma\Phi\epsilon I}{\lambda} (1 - e^{-\lambda t_i})(e^{-\lambda t_t})(1 - e^{-\lambda t_c}) \quad (9)$$

For a given cycle time T , the radiation response is maximized with $t_t = 0$ and $t_i = t_c = \frac{1}{2} T$. In other words, the transfer time is to be minimized while the irradiation period and counting period are to be the same length in order to obtain the most counts. However, that means response from background and other isotopes is maximized as well. Therefore, to optimize the cyclic system, both response in interest and response from background radiation are to be considered and compared. The quantity to be analyzed is the response to background ratio. This quantity is obtained in the following analysis.

Straight from equation 9, the counted response in the first cycle can be calculated by

$$D_1 = D = \frac{N\sigma\Phi\epsilon I}{\lambda} (1 - e^{-\lambda t_i})(e^{-\lambda t_t})(1 - e^{-\lambda t_c}) \quad (10)$$

The response in the second cycle is the total of activities from the second irradiation and the residual activities from the first cycle.

$$D_2 = D + D(e^{-\lambda T}) \quad (11)$$

Similarly, response from third cycle equals to

$$D_3 = D + D(e^{-\lambda T}) + D(e^{-\lambda 2T}) \quad (12)$$

And for the n th cycle we get

$$D_n = D \frac{(1 - e^{-n\lambda T})}{(1 - e^{-\lambda T})} \quad (13)$$

The total cumulative response D_c from all n cycles is obtained by summing up the measured responses.

$$D_c = \sum_{i=1}^n D_i \quad (14)$$

Equation 14 can be applied to activity response from both the isotope of interest and the response from background.

$_sD_c$ - cumulate response from isotope of interest;

$_bD_c$ - cumulate response from background radiation and other isotope in the specimen;

It was suggested to quantify the signal-to-background ratio in terms of the standard deviation of the background (Spyrou, *et al.* 1979).

$$R_{s-b} = _sD_c / \sqrt{ _bD_c } \quad (15)$$

The square root in equation 15 treats the background response as uncertainty into response from isotope of interest. It is possible to enhance the signal-to-background ratio by adjusting the number of cycles and decay time (Spyrou, *et al.* 1979). Figure 3 compares the theoretical signal-to-square-root-of-background ratio of a CNAA and a conventional one-time NAA. The ratio for both methods increases along with experiment time over the early stage. However, starting from about fourth minute, the ratio of the one-time NAA starts to drop due to the fading of responses from the short-lived isotope of interest. On the other hand, the ratio for CNAA keeps rising along with experiment time. The response from the isotope of interest does not fade out over time since the isotopes are irradiated again in each cycle. The isotope used to demonstrate in Figure 3 has an imaginary half-life 100 times shorter than that of the background radiation. If the half-life ratio increases to more than 100, both curves are expected to shift to the left; vice versa. A left-ward shift of the curve has two indications: 1) a favored reduced amount of total experiment time to achieve a desired R_{s-b} level; 2) a higher R_{s-b} ratio from short-lived isotopes over the same test period.

Figure 3. Signal-to-Background Ratio of Conventional and Cyclic NAA (Spyrou and Kerr 1979)

This proves CNAA a very effective analytical technique for very short-lived isotopes. In this research project, the cut “very short-lived” cut-off is half-lives of less than one minute.

2.3 EPITHERMAL NEUTRON ACTIVATION ANALYSIS

One major problem encountered using NAA is the dominance of radiation from isotopes with high cross-section such ^{27}Al , ^{23}Na , ^{37}Cl and ^{55}Mn due to their abundance in various types of specimens. The detector can be overwhelmed by the gamma activity and fail to detect the activity from other isotopes due to the very high backgrounds. One way to suppress the activities from these elements is by using epithermal neutrons with energy range between 0.5 eV and 500 eV, since they have very poor resonance cross-sections.

To understand the theory of ENAA, the first step is to determine the energy distribution of the neutron source. The neutron source employed for this research project is the TRIGA reactor installed at NETL. TRIGA reactor runs on TRIGA fuel, which is uranium-zirconium-hydride, UZrH (General-Atoms 2010). The reactor generates neutrons by fissions of ^{235}U . The energy spectrum of prompt neutrons produced by fission of ^{235}U can be estimated with expression

$$\chi(E) = 0.453 e^{-1.036E} \sinh \sqrt{2.29E} \quad (16)$$

$\chi(E)$ is the number of neutrons emitted at energy level E per fission per unit energy level. Energy level E is in unit of MeV. Plotting $\chi(E)$ versus energy level is illustrated in Figure 4.

Figure 4. Energy Distribution of Prompt Fission Neutrons

The distribution has a local maximum at 0.85 MeV, which is the most common neutron energy level. The mean energy level of all prompt fission neutrons can be calculated by integrating the product of energy level E and local probability across all energy levels. Solving the integration gives a mean energy level of 1.98 MeV (Lamarsh 2002).

$$\bar{E} = \int_0^{\infty} E \chi(E) dE = 1.98 \text{ MeV} \quad (17)$$

The reactor fuel, at which the prompt neutrons are born, is surrounded by a moderating medium. In a TRIGA reactor, light water is selected to be the moderator for a few reasons:

1) Water has a low neutron absorption cross-section at epithermal energies yet high scattering cross-section. 2) Water is a compound of hydrogen and oxygen. Hydrogen is the best neutron slowing element since it has an almost identical mass as a neutron. 3) Water can serve as a cooling agent at the same time. 4) Light water is easily accessible with low cost.

As neutrons pass through the water medium, they interact with water molecules. Due to the low neutron absorption cross-section, the dominant interaction with water is elastic scattering. Neutrons experience a series of collisions before they make their way out of the moderator medium into the irradiation chamber. Each time a neutron collides and scatters, part of the neutron energy is lost to the water molecule. Take an example of neutron born at energy E^* and is brought down to energy E after all the scattering. The likelihood of neutrons arriving at energy level E is proportional to $1/E$, for all starting energy E^* .

$$\chi(E) \propto 1/E \quad (18)$$

In other words, there are more neutrons in the low energy level range than the high energy level range. This agrees with the expectation that 90% of neutrons in a moderated neutron flux are at energy level below 1 eV (Shultis and Faw 2000). Under these conditions, the energy distribution of neutrons reaching the specimen is plotted as Figure 5.

Figure 5. Neutron Energy Distribution of Neutrons Reaching the Irradiation Chamber

Before the neutrons reach the specimens sent to the irradiation chamber, a cadmium target is present to absorb thermal neutrons. Referring to Section 1.4, the cadmium cutoff is at 0.5 eV. It is assumed that only neutrons with energy higher than 0.5 eV pass through the cadmium shield. However, this also means a large portion of neutrons are filtered away due to the trend that neutrons tend to exist at thermal energies

To illustrate the effective of epithermal NAA activation rate of aluminum and silver, respectively. Back from Figure 1, aluminum has an almost constant neutron cross-section of 0.5 barns across the energy range between 0.1 eV to 1000 eV. Meanwhile, silver has a resonance peak across 1 eV to 10 eV. Due to the $1/E$ trend, most neutrons to the specimens have energy range between 0.5 eV to 10 eV. As stated above, most neutrons are absorbed by the cadmium target. Therefore, the number of activated aluminum atoms is greatly reduced. On the other hand,

as shown in Figure 6, silver has a large absorption cross-section for the remaining neutrons, the reduction percentage of silver activities is not as high as that of aluminum. The reduction in activities is measured in cadmium ratio. The cadmium ratios of activities from ^{28}Al and ^{110}Ag are 48.5 and 2.39 respectively. In other words, by employing epithermal neutrons for the analysis, the radiation activity from aluminum is dropped by a magnitude of 48.5 while the activity from silver is cut by a magnitude of 2.39.

To conclude, ENAA is effective in identifying nuclides with resonance peaks across the epithermal range.

Figure 6. Neutron Total Cross-Section of ^{109}Ag (NNDC 2006)

3 Experimental

3.1 EXPERIMENTAL DESIGN

The purpose of this research project is to characterize the fast pneumatic transfer facility installed in the NETL for cyclic neutron activation analysis applications. Throughout this project, four separate conducted experiments were conducted.

1. Statistics of neutron flux source using aluminum wire specimens

The first experiment consists of irradiation and counting of 29 aluminum wire specimens, in order to determine the fluctuation of the neutron flux from the reactor. The standard deviation of the 29 measurements is used to calculate uncertainty for the subsequent experiments.

2. Determination of elements in biological and geological specimens

Biological and geological samples such as leaves and coal respectively were tested using CENAA to track short-lived isotopes if any, in attempt to demonstrate the usefulness of both epithermal neutrons and the cyclic approach.

3. Determination of elements in metallic ore specimens

CENAA is applied to track short-lived isotopes in irradiated metallic ore specimens. This experiment is to illustrate the effectiveness of NAA on metallic elements.

4. Determination of silver in air filters from the Arctic

There is interest in kinds of airborne particles that can be found in the Arctic. Sixty-five air filter samples collected across the year of 2009 were evaluated for elemental content using NAA. Silver was found in several samples and the element's trend is studied.

3.2 EXPERIMENT APPARATUS

The apparatus and facilities used in the research project are introduced in this section.

3.2.1 Fast Pneumatic Transfer Cyclic Irradiation Facility

In 2009 an automated pneumatic system was installed at NETL for the purpose of cyclic neutron activation analysis. The system can be divided into 4 parts, which are sample loading dock, transportation channel, counting chamber and control module. To operate the system, the operator inputs the needed parameters, which include irradiation time, decay time, count time, number of sample and number of cycles per sample, into the control module. Samples contained in vials are then loaded into a hopper, which is capable of holding 50 samples. An air pistol at the bottom of the hopper injects one vial at a time into an elevator, which sends the vial down to a network made of transportation pipes. A schematic and a picture of the loading dock are illustrated in Figure 7 and Figure 8 for a better understanding.

The pipe channel links the loading dock with irradiation chamber in the reactor core, germanium detector, and irradiated sample storage respectively. Since air contains radioactive argon which can activate to ^{41}Ar , carbon dioxide is purged to get rid of air within the channel before each operation. Once a vial enters the channel, an air electric pump generates a pressure difference and drives the vial to the irradiation chamber located in the reactor core. Inside the irradiation chamber, specimens are allowed to interact with neutrons emitted by reactor fuel. After the vial is irradiated for a desired period of time, the air pump generates a flow in the opposite direction and sends the vial near the germanium detector for counting measurements. For a one-shot NAA, the vial is immediately returned to the vial elevator, and is carried to a lower channel which leads to the disposal storage. In cyclic NAA, the vial is sent back and forth between the irradiation chamber and germanium detector for irradiation and counting before disposal. Used samples are set aside to allow the activities to decay before reuse or disposal. Multiple optical sensors are installed along the transportation channel to notify the system when a vial passes through. With the feedback from these sensors, the system is able to locate the vial

and execute a corresponding action of the electric pump and valve to achieve a precise irradiation time and count time for each of the cycles.

Figure 7. Loading Dock Sectional Diagram

Figure 8. Loading Dock

The counting chamber is the area that the germanium detector measures the activated specimens. The irradiated specimens are transferred to a spot next to the germanium detector probe. Gamma photons emitted along with beta decays of the activated nuclei are detected and converted into signals by the germanium detector. The signals are transmitted to a multichannel analyzer (MCA) which sorts the signals into different energy groups. Each energy group is represented by a channel. The frequency of signals from each energy group is plotted in terms of counts versus channel. The plot is known as a gamma spectrum. Spectra are displayed in the computer consecutively and are stored in database sorted by time signature. Since the activated specimens are radioactive, safety measures are required to protect the analysts and other individuals in the room. The counting area is surrounded by lead bricks to shield away gamma radiation from the activated specimens. Inside the lead cave cadmium and copper shields are used to eliminate the $k_{\beta I}$ x-rays from lead and copper. In addition, a GM tube is installed on top of the area to monitor the gamma exposure beneath the area. A picture of the chamber is show in Figure 9.

To summarize the roles of different parts of the system, a functional diagram of the automated system is shown in Figure 10.

Figure 9. Functional Diagram of the Automated Pneumatic System

Figure 10. Functional Diagram of the Automated Pneumatic System

Figure 11. An Overview of the Entire System

3.2.2 NETL TRIGA Reactor Facility

The neutron source during irradiation is the TRIGA Mark II reactor installed in NETL. TRIGA Mark II reactor was manufactured by General Atomics in 1986. The reactor is capable to generate a maximum of 1100 kW steady thermal power. The reactor has a light water moderator medium and a mix of water and graphite reflector. Five horizontal ports are installed for different experiments and research purposes (IAEA 2010). A neutron flux is generated in the core as NAA irradiation source. The intensity of the neutron beam can be adjusted through tuning the reactor power level. For epithermal NAA purposes, the power level is usually set to 100 kW or 500 kW, with a neutron flux of about 0.25×10^{11} and 1.2×10^{11} neutrons $\text{cm}^{-2} \text{s}^{-2}$ respectively.

3.2.3 High-Purity Germanium Detector (HPGe)

The gamma radiation measuring device used in this research project is a high-purity germanium detector. Germanium is classified as semiconductor. Electrons in semiconductors do not exist in a continuous energy range but in energy bands. Between any two adjacent energy bands, there is an energy gap called *band gap*. Electrons are not allowed to possess energy in the range of the band gaps. At equilibrium, each band allows certain number of electrons in its orbit. When an electron gains enough energy to overcome the band gap, the electron can jump over the band gap and reach a higher level band. Since there is only room for certain number of electrons in one band, the excited electron forces another electron in the band to travel to a neighboring band. The new electron enters the band and forces another electron out. On the other hand, the original band loses one electron and has a vacancy call a hole. An electron moves from a neighboring band in order to fill the hole. As a result, a hole is created in the neighboring band and is filled by electron from another band. The chain movements of the electron and the hole generate an electric current throughout the semiconductor. The currents are picked up as a measure of incident gamma ray and the electric potential of the current quantifies the energy level of the gamma radiation (Chen 2005).

Due to the delicate characteristic of the detection mechanism, it is important to have a pure germanium crystal for the detector. Any impurities may have a significant effect on the efficiency of the detector. Fortunately, the fabrication techniques which were developed in the last 40 years allow an impurity level as low as $10^9 \text{ atoms cm}^{-3}$, which is negligible compared to the atomic density of germanium, $4.41 \times 10^{22} \text{ atoms cm}^{-3}$.

3.2.4 PLC Com

All electronic devices, such as air pumps, valves and sensors installed in the pneumatic transfer facilities are controlled and monitored by computer program and integrated circuits. The computer program automatically runs through the entire experiment according to the time parameters, which are input by the user through the PLC Com portal. The panel of the portal, as shown in Figure 12, has boxes for irradiation time, count present, decay present, number of samples and number of cycles. The right hand side of the panel shows progress of the experiment. The center of the panel displays the gamma spectrum of the sample currently under counting.

Figure 12. PLC Com User Panel

Above the gamma spectrum, there is a row of buttons of “New”, “Start”, “Finish” and “Exit”. Detailed instruction to operate the pneumatic irradiation facility is given in Section 3.5.

3.2.5 Specimen Vials and Containers

Specimens are contained in low density polyethylene vials as they are transferred into and out of the reactor core.. The ethylene molecule is C_2H_4 ($H_2C=CH_2$)

while polyethylene is a polymer is a repetitive ethylene molecule depicted as

Carbon and hydrogen atoms have low neutron absorption cross-section and thus has a negligible contribution to measurement error. Other advantages include recyclability, transparent characteristic, low economical cost and availability of various sizes and shapes. There are three vial sizes used in this research project: 2-dram vials, 2/5-dram vials and micro vials, as shown in Figure 13. The 2-dram is the largest vial with 5 cm inner length and 1.5 cm inner diameter. The 2/5-dram is about half of the size of a 2-dram with 1.5 cm length and 1 cm diameter. And the micro vial has a 1 cm inner length and 0.5 cm inner diameter.

Specimens are first contained in either a micro vial or 2/5-dram vial, depending on the specimen size and weight. The small vials are then stored in 2-dram vials which are sent to the reactor by the pneumatic facility.

Figure 13. Polyethylene Vials; (from left to right) 2-Dram Vial; 2/5-Dram Vial; Micro Vial

3.3 SAMPLE PREPARATION

As discussed in section 1.2, samples are contained in vials which are carried by a carbon dioxide flow through a channel of pipes. In this research project, there are different types of samples in different physical forms such as solid, liquid and air filters. Therefore, it is essential to understand how the basic procedures samples are prepared and sealed in vials.

Most solid samples in this project are in powder form besides the air filters which are the standard Whatman filters. Sample powder, about 0.2 g, is first weighed in the micro vial and then placed into a 2/5 dram container. To do so, an empty micro vial is first placed on an electronic scale. The reading of the scale is then zeroed before loading any sample into the container. Protective gloves should be worn when unknown samples are handled. Transporting tools such as spoons should be cleaned before any contact with the samples. The vial should be filled no more than 90% full to leave room for the cap to close. A wipe should be used to clean any material on the outer surface of the container and on the edge of the opening, in order to get an exact weight measurement of the material inside. The scale reading is written down and the sample is assigned a number for future reference. Afterwards, close the cap of the dram carefully without letting any material spill out. An assigned number is marked on the side of the 2/5 dram with an oil based marker. The 2/5 dram containing the sample is then placed into a full size 2 dram vial. In this step, spacers are often used to keep sample in the middle of the vial and take out excess space to prevent impact during transportation.

Liquid samples are prepared in similar ways as solid samples are. Pipettes should be employed to control the volume of liquid in each batch of samples. After filling the desired volume of liquid into a micro vial the cap is closed and placed in a 2/5 dram vial and heat-sealed. For the air filters only the 2/5 dram vial was used. To check if the cap is fully sealed, the dram is submerged into a beaker of tap water. The dram is softly squeezed with a pair of tongs and observed carefully. The presence of bubbles indicates an incomplete seal. Sample liquid may leak and contaminate the facility. After the 2/5 dram is well sealed, it is placed in the middle of a 2 dram vial with spacers at either end.

Before the vials are allowed into the pneumatic system, they are placed through a test jig for a clearance test. The jig has a short piece of pipe with the same diameter as the pipes in the transportation channel. To perform the test, the vial is simply released and allowed to fall due to gravitation. A passing vial clears the pipe without any help of external forces while a failing vial is stuck in the pipe. An oversized vial may cause blockage in the transportation channel and may affect experimental time and data precision. Severe blockage may cause damage to the system; for example, a jammed vial with activation material may yield excessive radiation to the sample. Two common reasons for failing the jig test are 1) leftover polymer that sticks out at the bottom of the vial; 2) vial cap is not properly placed and enlarges the size of vial mouth. The samples are ready for loading once they pass the jig test.

A list of all the samples used in this work is presented in Table 5.

Table 5. Sample Contents and Purposes

Category	Content	Reference ID	# of Samples	Purpose
Aluminum	Aluminum Wire		30	Neutron Flux Statistics
Biological Samples (Reference Material)	Tomato Leaves	1573a	3	CENAA of Biological Samples
	Apple Leaves	1515	3	
	Peach Leaves	1547	3	
	Wheat Flour	1567	3	
	Rice Flour	1568a	3	
	Pine Needles	1575	3	
	Bovine Livers	1577b	3	
Coal Trace Elements	Coal Trace Elements	1632c	2	CENAA of Coal
Metal Ores (Canadian Standard Reference Materials)	Gold	CH-2	3	CENAA of Ore Samples
	Concentrated Zinc	CZN-1	3	
	Concentrated Lead	CPB-1	3	
	Nickel Copper Cobalt 1	SU-1	3	
	Nickel Copper Cobalt 2	UM-1	3	
	Nickel Copper Cobalt 3	SU-1A	3	
	Nickel Copper	PTM-1	3	
	Zinc Lead Tin Silver	KC-1	3	
	Zinc Tin Copper Lead	MP-1	3	
Arctic Samples	Air Filters		65	CENNA of Airborne Elements in the Arctic
Silver Reference	Silver Solution		4	Concentration Reference of Silver

3.4 SETUP OF THE PNEUMATIC TRANSFER SYSTEM

Before any samples are loaded into the fast pneumatic system, a number of checkups should be covered to ensure safety and performance.

3.4.1 Completion of Transfer Circuit

It is necessary to complete the transfer circuit connecting the reactor core and the fast pneumatic system. This is done because there is only one route that enters the irradiation chamber and the route is shared with another transfer mechanism, which is operated along with manual sample handling. There are two junctions to be adjusted to switch from one transfer mechanism to another. The first one is located on the third floor ceiling of the reactor bay, outside of the sample handling room. The joint is in form of a cylindrical revolver, in where the inlets of the air pump are planted. A lever is to adjust the angle of the revolver and the connection with either one of the two transfer mechanisms. The second connecting point is located on top of the reactor. One of the two outlets connects to the passage leading to the irradiation chamber and the joint is secured with a screw mechanism.

3.4.2 Carbon Dioxide Supply

As discussed in section 3.1, carbon dioxide is filled into the channel to replace air as the transfer fluid due to the presence of ^{41}Ar in air. Therefore, it is important to ensure a sufficient supply of carbon dioxide for the experiment. The source of carbon dioxide is a gas tank connected to the transfer channel through a gas valve. A barometer is installed at the valve to monitor the pressure of the carbon dioxide in the container. The pressure stays at about 800 psi before the gas runs out and requires replacement. It is noted that the valve is left open through the whole experiment in order to generate a pressure slightly higher than the atmosphere to prevent backflow of air into the system.

3.4.3 Position of Germanium Detector

The distance between the germanium detector and the radioactive target is an important factor that contributes to the efficiency of the germanium detector. This is because the gamma flux entering the detector is very sensitive to distance. To explore the correlation, consider both a point detector and an isotropic point source that emits photons in all directions. Gamma flux at the point detector can be calculated with the following expression.

$$\Phi_o = S/4\pi r^2 \quad (19)$$

where Φ_o - photon flux, in units of photons per unit area per second, without air attenuation

S - photon emission rate, in photons per second

r - distance between source and detector

The term $4\pi r^2$ is the area of a plane, on which all points have distance of r from the point source. Since the source is isotropic in all directions, the photon flux is identical on any point of the plane. Therefore, the photon flux is calculated by dividing the photon emission rate with the area of the plane. If the source emission rate is considered a constant, the flux equals to a product of constants and $1/r^2$.

$$\Phi_o \propto 1/r^2 \quad (20)$$

The expression illustrates the inverse proportional correlation between radiation flux and square of distance from source. This correlation is known as the inverse-square law or simply $1/r^2$ law.

The distance between the detector and the sample should be adjusted before each experiment. It is possible to move the detector through a digital device which sits next to the detector and controls a motor that drives the detector back and forth in a single yaxis movement. However, there are no measurement devices that keep tracks of the distance. Instead, the location of the detector is marked with a piece of paper wrapping the neck of the air chamber, for the purpose of experiment reproduction.

3.4.4 Reactor Startup and Power Adjustment

The TRIGA reactor in NETL is operated by experienced reactor operators. They are responsible for reactor start-up, power adjustment and shutdown. It is their duty to ensure the reactor functioning properly and safely. Therefore, the operators should be acknowledged before any actions are done or samples are sent into the reactor.

For any emergency the reactor operator is informed to scam the nuclear reactor.

3.4.5 Test Run

It is strongly suggested by the reactor operators at NETL that a blank sample vial should go through the whole system before any specimens are loaded. The purpose of the dry run is to ensure the clearance of the channel passages and proper functioning of all the valves. Other than a blank sample, a sulfur powder specimen, which has a very low activity of activation radiation, can be used instead of a blank to test the presence of neutron beam. The suggested input parameters for a test run is 10 second irradiation time, 10 second decay time and 10 second counting time run for one cycle.

3.5 Experimental Procedure

Since the fast pneumatic transfer cyclic irradiation facility is fully automated, the experiment is regarded as instrumental experiment. The general procedure is identical regardless of the different sample types.

1. Load samples into the sample hopper. Make sure all the vials are closed with caps. The opening ends of the all vials should face the direction of the germanium detector. Such orientation allows the open end face upwards, in order to prevent any leakage of material inside the reactor core through the opening.
2. Open the PLC Com panel. Click on “New” for a new set of time parameters. The action should also zero all the input squares from the previous experiment. After keying in the irradiation time, count present, decay present, number of samples and number of cycles, hit

“OK” at the bottom right corner. All input parameters should be displayed on the status screen next to the computer. Do not hit the “Start” button yet.

3. Inform the reactor operator about the incoming of samples with the intercom in the room. This is important as the reactor operators are responsible to keep track of all the materials going into the reactor core. The reactor operator will respond and hit the “enable” key in the control room when he/she finds the reactor ready.

4. When both “Start” and “Enable” are activated, the air pump starts purging carbon dioxide gas into the transfer channel. This progress should take about 5 minutes before the first sample is loaded and sent into the channel.

5. Samples are sent into the core for irradiation and are brought back to the germanium detector for counting. Once the counting is finished, the data is saved in the “user” folder under the day’s date in C: drive of the computer, in the form of “finish time_sample number.chn”. For example, if count for the fifth sample is finished at 2:20 pm, the file will be “1420_005.chn”. chn files can be opened and viewed using a software called Maestro.

6 Counted specimens are ejected from the facility and are stored in a lead shielded bucket near the facility. The specimens should be idled for a few days before they are handled and reused.

7. Repeat from step 1 with multiple samples. Inform the reactor operators when finished.

4 Results and Discussion

There are four performed experiments in total for this research project. Results from each of the experiments are discussed below.

4.1 NEUTRON FLUX STATISTICS

Each of the 29 aluminum wire specimens was irradiated for 10 seconds in the reactor core at power of 100 kW before they are sent to the germanium detector. The counting progress starts exactly 10 seconds after the specimen leaves the irradiation chamber in the reactor core. A gamma spectrum was recorded for of 60 seconds. A typical spectrum is seen in Figure 14. The primary gamma-ray of 1778.9 keV comes from the $^{27}\text{Al}(n,\gamma)^{28}\text{Al}$ reactions. The gamma-rays of 843.8 keV and 1014 keV belong to the $^{27}\text{Al}(n,p)^{27}\text{Mg}$ reaction.

More often the 511 keV photons are created by pair production followed by annihilation. An incident photon is absorbed and its energy is converted into a positron-electron pair. Since the rest mass of electron and positron is 511 keV, the threshold energy for pair production is 1022 keV, or 1.022 MeV. The positron born in pair production interacts with an electron in terms of annihilation. The rest mass of the two particles is converted into energy and creates two photons each at 511 keV.

Figure 14. Gamma Spectrum of an Aluminum Wire

For statistical purposes, the only peak of interest is the 1778.8 keV. The net area of the peak, or the total counts at and can be obtained with Maestro MCA. The net area values of each of the 29 specimens are imported onto a spreadsheet for further calculation. The values are first normalized by the specimen weight in terms of net area per unit weight, as donated by equation 21.

$$A_{wi} = \text{net area about peak at 1778.8 keV} / \text{specimen weight} \quad (21)$$

The average of the A_w of all 29 specimens can be easily found with equation 22.

$$\overline{A_w} = \sum_{i=1}^{29} A_{wi} / 29 \quad (22)$$

The density distribution of the aluminum wires is assumed to be isentropic. In other words, there is identical number of aluminum atoms in each unit weight of wire. It is also assumed that each activated nucleus has the identical chance to emit a gamma particle that is picked up by the detector. With these two assumptions, the only variable factor that contributes to the variation of the net area about the gamma spectrum peak is the neutron flux impinging onto the aluminum specimens.

The next step is to normalize the data by this average and generate a unit-less value.

$$I_i = A_i / \bar{A_w} \quad (23)$$

And the unit-less value is put into a line plot in Figure 15.

Figure 15. Fluctuation of Neutron Flux

Since the data is normalized, the value of 1 represents the average of the 29 data points. The plot illustrates the maximum distance from the average is about 0.074 or 7.4%. And the 29 points span a maximum difference 0.141 or 14.1%. To better quantify the potential error, the standard deviation of these 29 points can be calculated by equation 24.

$$\sigma = \sqrt{\sum_{i=1}^{29} (I_i - 1)^2 / (29 - 1)} \quad (24)$$

The standard deviation equals to 0.041 or 4.1%, which means there is a 4.1% standard deviation from the neutron flux. The result is used in calculation of uncertainty for the subsequent experiments.

4.2 BIOLOGICAL AND COAL SAMPLES

The first approach of CENAA was to analyze biological samples. The specimens include NIST certified reference materials of tomato leaves, apple leaves, peach leaves, wheat flour, rice flour, pine needles and bovine liver. It is expected that the major content of these specimens are organic substances which do not have a high neutron absorption cross-section.

Figures 16 to 22 show the spectra of the seven specimens that undergo 5 cycles, each cycle with 10 seconds of irradiation, 10 second decay and 30 seconds of counting. The major peaks of the spectra are labeled with their corresponding gamma energy in keV and the isotope from which the gamma ray is emitted from.

Figure 16. Gamma Spectrum of Tomato Leaves

Figure 17. Gamma Spectrum of Apple Leaves

Figure 18. Gamma Spectrum of Peach Leaves

Figure 19. Gamma Spectrum of Pine Needles

Figure 20. Gamma Spectrum of Rice Flour

Figure 21. Gamma Spectrum of Wheat Flour

Figure 22. Gamma Spectrum of Bovine Liver

It is within expectation that gamma activity from ^{28}Al contributes significantly to the gamma spectra due to its high natural abundance and high decay rate. With the removal of thermal neutrons in the neutron flux, the activity rate from aluminum atoms is significantly suppressed and allows detection of other isotopes. Comparing the peak height of ^{28}Al and other major isotopes, the peak ratio is within the range of 10 to 100.

Isotopes found among the biological samples include common metallic elements such as aluminum, sodium, magnesium and manganese. Non-metallic elements include bromine, iodine, chlorine and oxygen. Table 6 lists the isotopes found on the spectrum and in order of their half-lives

Table 6. Elements in Biological Samples (in order of half-life)

Isotope	Gamma Energy (keV)	Half-life
¹⁹ O	197.4, 1356.9	26.1 sec
²⁸ Al	1778.8	2.24 min
²⁷ Mg	843, 1014	9.46 min
⁸⁰ Br	511, 617	17.68 min
¹²⁸ I	442.9, 526.6	24.99 min
³⁸ Cl	1642, 2167.6	37.24 min
⁴¹ Ar	1293.6	1.83 hr
⁵⁶ Mn	846.8, 1810, 7, 2113	2.58 hr

The result is disappointing having ¹⁹O being the only isotope with a half-life shorter than one minute among the isotopes found in the biological specimens. Therefore, in order to illustrate the effect of the cyclic approach, it was decided to investigate other types of specimens in search for other short-lived isotopes under one minute.

An available option was to determine short-lived isotopes in coal. The gamma spectrum obtained from the specimen is shown in Figure 23.

Figure 23. Gamma Spectrum of Coal Trace Element

A similar result was obtained from coal as those from biological specimens. The spectrum illustrates the presence of aluminum and magnesium.

4.3 METALLIC ORE SPECIMENS

Metallic ores were selected as the next approach to find short-lived isotopes that benefit from both epithermal neutrons and cyclic NAA technique. The activated contents in the ores generate a much larger amount of radiation activity than those in biological specimens. Therefore, the ore specimens were irradiated under a lower reactor power, 20-50 kW with larger detector spacing relatively to the previous experiment configuration. The specimens were irradiated for 10 seconds and counted for 30 seconds after a 10 second delay in each cycle. Nine ore specimens from seven ore species were analyzed and their gamma spectra are shown in Figures 24 to 30. Only one of the three nickel-copper-cobalt ore spectra is shown due to their similarity.

Figure 24. Gamma Spectrum of Gold Ores

Figure 25. Gamma Spectrum of Nickel-Copper Ore

Figure 26. Gamma Spectrum of Nickel-Copper-Cobalt Ore 1

Figure 27. Gamma Spectrum of Zinc-Lead-Tin-Silver Ore

Figure 28. Gamma Spectrum of Zinc-Tin-Copper-Lead Ore

Figure 29. Gamma Spectrum of Concentrated Lead Ore

Figure 30. Gamma Spectrum of Concentrated Zinc Ore

Compared with the spectra obtained from the biological and coal specimens, there are two significant differences observed from the metallic ore spectra. The first observation is that ^{28}Al is no longer the dominant activity source for many of the specimens. Another observation is that the measurement dead time is much higher than that with biological specimens. Possible countermeasures are decreasing the neutron flux or increasing the distance between the specimens and the detector probe. Further discussion about dead time is made in Section 4.5 and 4.6.

Table 7 lists the major isotopes found within the nine ore specimens.

Table 7. Elements in Metallic Ore Specimens (in order of half-life)

Isotope	Gamma Energy (keV)	Half-life
^{110}Ag	657.8	24.6 sec
^{28}Al	1778.8	2.24 min
^{52}V	1434.1	3.75 min
^{66}Cu	1039.2	5.1 min
^{27}Mg	843, 1014	9.46 min
$^{60\text{m}}\text{Co}$	58.6, 1332.5	10.47 min
$^{116\text{m}}\text{In}$	416.9, 818.7, 1097.3, 1293.5, 2122.1	54.15 min
^{56}Mn	846.8, 1810.7, 2113	2.58 hr

As expected, majority of the short-lived isotopes found in ores are from metals. The most short-lived isotope is ^{110}Ag with a half-life of 24.6 seconds, which is under the cutoff of one minute. Furthermore, referring back to Figure 6, ^{109}Ag has a strong resonance neutron absorption cross-section peak over the epithermal range of 1 eV to 10 eV. So far to this point of the research project, ^{109}Ag is the first isotope that demonstrates both criteria for CENAA. Therefore, silver is chosen as the element of interest.

It is remarked that the gamma peak at 1267 keV is not result from activated silicon nuclides. Instead, the 1267 keV photons are from ^{28}Al , which has an energy level of 1778 keV,

less the rest mass of electron, 511 keV; i.e. $1778 \text{ keV} - 511 \text{ keV} = 1267 \text{ keV}$. This is the single escape peak.

To illustrate the advantage of cyclic NAA over a one-time conventional NAA, two zinc-lead-tin-silver ore specimens of the same mass were irradiated and counted for one and two cycles, respectively. The signal-to-background ratio, R_{s-b} , is calculated using equation 25.

$$R_{s-b} = {}_sD_c / \sqrt{{}_bD_c} \quad (25)$$

According to the gamma spectrum of zinc-lead-tin-silver ore, the major activities picked up by the detector are from silver and aluminum. With silver as the isotope of interest, activities from ^{27}Al represent background radiation.

${}_sD_c$ = detector response from isotope of interest

= net area about the peak at 657.8 keV, which is the gamma energy from ^{110}Ag

${}_bD_c$ = detector response from background radiation

~ detector response from aluminum

= net area about the peak at 1778.8 keV, which is the gamma energy from ^{28}Al

Obtaining the values from the two spectra and the calculated result is shown in Table 8.

Table 8. Signal-to-Background Ratio for One and Two NAA Cycles of Zn-Pb-Sn-Ag Ore

	1 cycle	2 cycle	
${}_sD_c (^{110}\text{Ag})$	12162	25107	
${}_bD_c (^{28}\text{Al})$	1607	4193	
			% Increased
R_{s-b}	303.3871	387.7327	27.80132

There is a 27.8% increase in signal-to-background ratio with a second cycle. The increase indicates the effectiveness of cyclic NAA in enhancing the signal of short-lived isotope activities.

4.4 Arctic Air Filters

CENAA is proven in the prior experiments to be an effective technique in determining short-lived isotopes with strong neutron resonance over the epithermal range. There has been interest in the airborne particle distributions in the Arctic Circle. Sixty five air filter samples were received from the Air Quality Research Division of Environment Canada. The air filters are collected in the Canadian side of the Arctic Circle, on a weekly basis and spanning over the year of 2009. Each specimen contains 1/16 of an air filter.

4.4.1 Ag Concentrations

Each of the 65 samples underwent CENAA of three cycles. Two of the 65 spectra are illustrated in Figure 31 and 32, which belong to two filters taken at the beginning of January and July respectively.

Figure 31. Gamma Spectrum of Arctic Air-Filter (Taken on January 7, 2009)

Figure 32. Gamma Spectrum of Arctic Air-Filter (Taken on July 7, 2009)

Comparing the two spectra, different elements are found within the Arctic atmosphere over different periods of time in the year. Figure 31 shows the presence of aluminum, argon and oxygen in January; while Figure 32 gives proof of presence of silver, tin, copper, and aluminum in the summer.

The presence of multiple metallic elements in the Arctic Circle suggests air flow from Northern Canada, where ore mines are located. Figure 33 shows a backward trajectories model that tracks the air flow towards the location where the airborne data was collected. The lines indicate the calculated route of air flow that reaches the location of the air sampler.

Figure 33. Backward Trajectory Model of Arctic Air Flow (Hysplit 2010)

4.4.2 Airborne Silver Concentration and Trend over Time

The silver net area counts (for 1/16 of an air filter) of the air samples are plotted against the date the samples are collected at. The air concentrations are derived by multiplying the concentration by 16 and dividing by the total air flow for the week.

Figure 34. Airborne Silver Concentration in the Year of 2009

Figure 35 shows the variation of airborne silver concentration across the year. Based on the result, the maximum silver content is 1.52 ng m^{-3} . The sample was collected in the week of July 7, 2009. The procedure to calculate the concentration is shown in the next section.

It was surprising to find an accumulating concentration over the period between the end of May and the beginning of August. Typically heavy metal concentrations in the Arctic appear during the winter months. It is also interesting to notice a sudden drop of silver concentration in mid-August. Another remarkable finding is the trend comparison between silver and aluminum, which is illustrated in Figure 35. The two metallic elements do not share any common trend over

the period of summer. The absence of correlation suggests the two elements were from different sources and were brought together by wind in different directions.

The finding of trends is worth further investigation and research. However, it will be far beyond the scope of this research project to explore the reason behind the trend.

Figure 35. Silver and Aluminum Trend over Summer of 2009

4.5 SILVER STANDARD REFERENCE AND CONCENTRATION CALCULATION

The final stage of the research project is to justify the accuracy of CENAA.

According the result from prior experiments, silver is found in a number of the air filters and ore specimens. The next approach is to determine the silver concentration in each of these samples. The idea is to perform CENAA with reference samples, of which the silver concentration is known, under the exact configurations as the prior experiments. The reference employed for this experiment is silver standard reference solution. The labeled silver concentration of the solution is 10 parts per million (ppm). Each of the reference specimens is filled with 0.1 g of the silver solution. Therefore, the mass of natural silver in each specimen equals to $0.1 \text{ g} \times 10/1000000 = 1 \mu\text{g}.$ The solution is irradiated and counted under the same reactor power and distance configuration as the prior experiments. Each specimen undergoes 3 cycles of irradiation and counting.

Table 9. Net Area Counts of Standard Reference Solution

Reactor Power (kW)	# of Cycles	Net Area (counts)	Uncertainty (counts)	Position	Mass (g)
20	3	129	15	B	0.1
50	3	343	23	B	0.1
100	3	642	33	B	0.1
500	3	7229	99	C	0.1

Table 9 lists the 4 different reactor power levels and number of cycles. The result is, as expected, showing a proportional correlation between reactor power and net peak area. Given the reference data and the net area count of a specimen, it is possible to calculate the unknown concentration of the specimen with the following quantitative approach.

A few assumptions are made to this approach. Firstly, it is assumed that all silver found in different specimens do have the same natural silver isotope abundance ratio; i.e. 52% of ^{107}Ag

and 48% of ^{109}Ag . Secondly, all ^{109}Ag atoms have identical neutron absorption cross-section and likelihood to be activated to emit a gamma particle of 657 keV. The two assumptions suggest that if two different specimens, under the same irradiation and counting circumstances, have the same net area about the 657 keV peak in their gamma spectra, then the mass content of silver in these two specimens are identical. Furthermore, the two specimens have the same net area to mass ratio. This correlation can be donated by the following equation.

$$C_i/M_i = C_{ii}/M_{ii} \quad (26)$$

where C_i and C_{ii} are the net area counts of specimen 1 and 2 respectively;

and M_i and M_{ii} are the mass content in specimen 1 and 2 respectively.

Equation 17 can be rearranged to have M_{ii} on one side and the rest of the variables on the other.

$$M_{ii} = M_i \times C_{ii}/C_i \quad (27)$$

Assuming specimen 1 is the silver standard reference and specimen 2 is the specimen of interest with unknown mass content. Equation 28 can be written in following form.

$$M_{uk} = M_k \times C_{uk}/C_k \quad (28)$$

The net area counts are across a 30 seconds live time spam instead of real time spam. The difference between real time and live time is defined as dead time, which is the detector blind time due to overflow of radiation activities. Each specimen has a different emission rate and thus a different time for each of the spectra.

One common countermeasure is a zero-dead-time (ZDT) correction modeling by the MCA. Unfortunately, dead time correction program for the fast pneumatic system currently is currently under development. There are two alternative options: using preexisting dead time equation and equation from other researchers or developing a new approach.

One existing model is introduced in “Neutron Activation Analysis” (Soete, Gijbels and Hoste 1972). Soete suggests

$$c/c' = \lambda t_d / (\exp(\lambda t_d) - 1) \quad (29)$$

where c' is the net area count with dead time correction;

c is the net area count obtained from the spectrum;

t_d denotes dead time;

λ is the decay constant of the isotope.

Another model that is derived by the team is based on the assumption that all dead time is caused by the activities of the isotope of interest and the likelihood of dead time occurring at time t is proportional to the activity distribution of the isotope.

$$f(t) = C \exp(-\lambda t) \quad (30)$$

Applying the boundary condition

$$F(t_{\text{real}}) = \int_0^{t_{\text{real}}} C \exp(-\lambda t) dt = 1 \quad (31)$$

gives $C = \lambda / (1 - \exp(-\lambda t_{\text{real}}))$.

The fraction of signals lost over dead time is derived by

$$C_{\text{loss}} / C_{\text{total}} = (\text{dead time fraction}) \int_0^{t_{\text{real}}} f(t) \exp(-\lambda t) dt \quad (32)$$

Substitute equation (30), constant C , and dead time fraction = dead time / real time

$$C_{\text{loss}} / C_{\text{total}} = \lambda t_d / [t_{\text{real}} (1 - \exp(-\lambda t_{\text{real}}))] \int_0^{t_{\text{real}}} \exp(-2\lambda t) dt \quad (33)$$

Deriving the integration gives

$$C_{\text{loss}} / C_{\text{total}} = t_d (1 - \exp(-2\lambda t_{\text{real}})) / [2t_{\text{real}} (1 - \exp(-\lambda t_{\text{real}}))] \quad (34)$$

Therefore, the net area count with dead time correction

$$c' = c / (1 - C_{\text{loss}} / C_{\text{total}}) \quad (35)$$

The results of the silver concentration calculation in ores based on the two estimation models are compared with the reported concentration in Table 10. In general, both dead time models project estimations on the silver concentration within 10% difference from the reported concentration.

Table 10. Silver Concentration in Ores Specimens

	w/o correction	Soete Model	New Model	Reported
Gold	22.24	22.53	23.06	24.2
+-	4.97	5.03	5.15	
Zn Pb Sn Ag	893.80	1036.26	1166.98	1120
+-	38.80	44.98	50.66	
Zn Sn Cu Pb	47.82	55.61	62.66	59.5
+-	17.26	9.86	11.11	
Ni Cu	73.92	77.05	80.86	66
+-	5.75	6.00	6.29	
Zn Conc	87.73	90.62	93.94	93
+-	6.67	6.89	7.14	
Ni Cu Co	4.57	4.68	4.79	4.3
+-	7.55	7.73	7.91	
Pb Conc	530.49	625.31	705.54	626
+-	23.63	27.86	31.43	

A similar approach is employed to determine the concentrations of the air filters.

The calculation results show that CENAA is effective in measuring element silver concentration in the Arctic air filters.

4.6 SOURCE OF UNCERTAINTY AND ERROR

Uncertainty estimation is necessary to justify the accuracy and precision of the silver content calculations. The total error percentage is the square-root of sum of error percentage square of each individual error source.

$$E_{\text{total}} = \sqrt{\sum_{i=1} E_i^2} \quad (36)$$

The major source of error to the silver concentration calculation in ores is dead time. Dead time refers to the short time flame after the MCA amplifier converts a signal. During this period of time the MCA is under a recovery process and is unable to record another incoming signal. As a result, a portion of signals are never recorded and the generated spectrum underestimates the actual activities. The fast pneumatic system in NETL is programmed to compensate the dead time by adding extra counting time. However, such countermeasure does not work well for short-lived isotopes since the decay rate over the compensation time is much lower than that overall dead time.

The major source of uncertainty in airborne calculation is the net peak area of the gamma spectrum. The low mass of target isotopes allows a high uncertainty-to-net-area ratio. The ratio can be suppressed by increasing the activity rate from the specimens. However, a higher activity rate would result in higher dead time.

Another possible source of error is gamma-ray self-attenuation., particularly for ores which contain high-Z elements. In other words, the specimen itself acts as a shielding medium and reduces the number of photons that make their ways to the detector. The self-attenuation effect is estimated by using a program called SELFABS (Dodoo-Amoo and Landsberger 2001), which calculates the fraction of photons that are attenuated by the major elements in the specimens. Table 11 lists the result.

Table 11. Photon Attenuation Percentage in Different Ore Specimens

	Average mass (g)	Major constituent	Attenuated %
Gold	0.230	Fe 25.7%, Cu 2.43%	1.100
Zn Pb Sn Ag	0.360	Zn 20.07%, Pb 6.87%	1.400
Zn Sn Cu Pb	0.280	Zn 16.33%, Sn 2.5%, Cu 2.15%	1.000
Ni Cu	0.460	Ni 47.77%, Cu 24.96%	3.570
Zn Conc	0.350	Zn 44.74%, S 30.2%, Fe 10.93%, Pb 7.45%	2.700
Ni Cu Co	0.250	Ni 1.233%, Cu 0.947%	0.560
Pb Conc	0.490	Pb 34.74%, S 17.8%, Fe 8.43%, Zn 4.42%	4.800

The concentration calculations are corrected by accounting the lost fraction of photons with equation 37.

$$c' = c / (1 - f_a) \quad (37)$$

where c is the concentration estimate before correction

c' is the concentration estimate after correction

f_a is the attenuated photon fraction

5 Recommendations and Future Work

A number of improvement measures can be made to enhance the efficiency of the analytical technique used in this research project.

5.1 DEAD-TIME CORRECTION

As mentioned in Section 4.6, dead time is the major source of experimental error. The dead time correction function will benefit by giving a more accurate gamma spectrum analysis.

5.2 SAMPLE TRANSFER TIME

The suggested decay time is 10 seconds since pneumatic system takes a minimum 8 seconds to bring the irradiated specimen from the reactor core to the counting chamber. For ^{110}Ag , with a half-life of 24.6 seconds, a 10 second decay time takes away $(1 - e^{-\lambda t}) = 24.36\%$ of its initial activity. On the other hand, the 10 second delay suppresses activity from ^{28}Al by 5.03%. That means in the beginning of the counting period, there are 75.44% and 94.97% of their activated activities for the two isotopes respectively. The source-to-background constant can be calculated with equation 29.

$$C_{s-b} = \exp(-\lambda t)_s / \sqrt{\exp(-\lambda t)_b} \quad (37)$$

Notice equation 37 is similar to equation 25 of signal-to-background ratio R_{s-b} which quantifies the detectability of the isotope of interest. The constant C_{s-b} is proportional to R_{s-b} and is the contribution factor of decay time. Table 12 lists the value of the constant at decay time of 0-20 seconds. A 10 second decay time gives a constant of 0.77, which implies a 23% reduction in the signal-to-background ratio R_{s-b} ; while a 6 second decay time reduces 14% of the ratio.

One possible configuration is increasing the flow velocity of the transferring gas in the ducts. However, the high velocity may generate a large amount of braking force and may break the cap of the vials, leaving the specimens in the transfer channel.

Table 12. Signal-to-Background Constant with Decay Time Below 20 Seconds

	Activity Fraction		
Decay Time	¹¹⁰Ag	²⁸Al	C_{s-b}
0	1.00	1.00	1.00
2	0.95	0.99	0.95
4	0.89	0.98	0.90
6	0.84	0.97	0.86
8	0.80	0.96	0.81
10	0.75	0.95	0.77
12	0.71	0.94	0.74
14	0.67	0.93	0.70
16	0.64	0.92	0.66
18	0.60	0.91	0.63
20	0.57	0.90	0.60

6. Conclusions

In conclusion, CENAA is an effective technique in determining short-lived ($T_{1/2} < 1$ minute) nuclides with high resonance peak in the epithermal range. However, it is disappointing to have ^{109}Ag as the only isotope that satisfies both criteria for a variety of reference materials. Moreover, CENAA application is well demonstrated in airborne silver determination in Arctic air filters.

The fast pneumatic transfer facility in NETL is still under its development stage. Gamma spectra and concentration calculations are expected to be more accurate upon the implementation of the ZDT software.

BIBLIOGRAPHY

- Barthelmy, David. *Mineral Species containing silver (Ag)*. 12 31, 2009. <http://www.webmineral.com/chem/Chem-Ag.shtml> (accessed 04 2010).
- Chatt, A., H. S. Dang, B. B. Fong, and C. K. Jayawickreme. "Determination of Trace Elements in Food by Neutron Activation Analysis." *Journal o Radioanalytical and Nuclear Chemistry*, 1988.
- Chen, Gang. *Nanoscale Energy Transport and Conversion*. New York: Oxford University Press, 2005.
- Dodoo-Amoo, D, and S Landsberger. "Gamma-ray self attenuation calcultions in neutron activation analysis: A problem Overlooked." *Journal of Radioanalytical Chemistry Vol. 248*, 2001: 327-332.
- Elayi, A.G., P. Damiani, and P. Collet. "14 MeV Neutron Activation Analysis of Gaulish Silver Alloyed Coins." *Journal of Radioanalytical and Nuclear Chemistry Vol. 90*, 1985: 113-122.
- Frame, Paul W. *A story about the "discovery" of neutron activation analysis*. 1997. <http://www.ora.u.org/PTP/articlesstories/fermi.htm> (accessed February 2010).
- General-Atomics. *About TRIGA*. 2010. <http://www.ga-esi.com/triga/about/index.php> (accessed March 2010).
- Glascok, Michael D. *Overview of NAA*. University of Missouri. 2010. http://archaeometry.missouri.edu/naa_overview.html (accessed 2010).
- IAEA. *Research Reactor Details - TRIGA II UNIV. TEXAS*. Februray 17, 2010. http://www-naweb.iaea.org/naweb/physics/research_reactors/database/RR%20Data%20Base/datasets/report/United%20States%20of%20America%20%20Research%20Reactor%20Details%20-%20TRIGAIUNIV_TEXAS.html (accessed March 2010).
- KAERI. Nuclear Data Evaluation Lab, Korea Atomic Energy Research Institute. 2000. atom.kaeri.re.kr (accessed 2010).
- Kruger, Paul. *Principles of Activation Analysis*. Wiley, 1971.
- Lamarsh, John R. *Introduction to Nuclear Reactor Theory*. American Nuclear Society, 2002.
- Mcdowell, L. S., P. R. Giffen, and A. Chatt. "Determination of Selenium in Individual Food Items Using short-Lived Nuclides." *Journal of Radioanalytican and Nuclear Chemistry Vol. 110*, 1987: 519-529.
- NETL. *Neutron Activation Analysis*. <http://www.me.utexas.edu/~netl/NeutronActivationAnalysis.htm> (accessed 2010).

- NNDC. *Sigma Plots of Cadmium-131 neutron total cross section*. 2006.
<http://www.nndc.bnl.gov/sigma/getPlot.jsp?evalid=4365&mf=3&mt=1&nsup=10>
 (accessed February 2010).
- . *Sigma Plots of Silver-109 neutron total cross section*. 2006.
<http://www.nndc.bnl.gov/sigma/getPlot.jsp?evalid=4357&mf=3&mt=1&nsup=Neutron%20reactions>
 (accessed 2010).
- Shultis, J. Kenneth, and Richard E. Faw. *Radiation Shielding*. La Grange Park: the American Nuclear Society, 2000.
- Soete, D., R. Gijbels, and J. Hoste. *Neutron Activation Analysis*. Wiley-Interscience, 1972.
- Spyrou, N. M. "Neutron Activation Analysis Challenges." *Journal of Radioanalytical and Nuclear Chemistry Vol. 239*, 1999: 59-70.
- Spyrou, N. M., and S. A. Kerr. "Cyclic Activation: The Measurement of Short-lived Isotopes in the Analysis of Biological and Environmental Samples." *Journal of Radioanalytical Chemistry Vol. 48*, 1979: 169-183.
- Spyrou, N. M., CH. Adesanmi, M. Kidd, L.G. Stephens-Newsham, A.Z. Ortaoval, and F. Ozek. "Usefulness of Thermal and Epithermal Cyclic Activation Analysis with a Reactor System." *Journal of Radioanalytical Chemistry Vol. 72*, 1982: 155-182.
- Taylor, Denis. *Neutron Irradiation and Activation Analysis*. D. Van Nostrand Company, Inc, 1964.

Vita

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