Twenty Five Years of Neutron Activation Analysis: A Personal Perspective on Utilization of the Techniques

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NAA Facilities

- As of 2010, there are 236 research reactors worldwide and another 6 under construction.
- Neutron activation analysis (NAA) still remains the most used technique in these facilities.

Interferences in NAA

- The use of thermal neutrons is the mainstay of NAA for all the facilities.
- The presence of aluminum, chlorine, sodium and manganese can inhibit the determination of several short-lived radionuclides due to increased backgrounds from the various reactions: ²⁷Al(n,γ)²⁸Al, ³⁷Cl(n,γ)³⁸Cl, ²³Na(n,γ)²⁴Na and ⁵⁵Mn(n,γ)⁵⁶Mn.

Interferences in NAA

- For medium-lived NAA the presence of ²³Na(n,γ)²⁴Na and ⁸¹Br(n,γ)⁸²Br can also greatly add to the Compton continuum to the point that elements such as arsenic need to be determined by radiochemistry.
- Reactions of ⁴⁵Sc(n,γ)⁴⁶Sc and ⁵⁹Co(n,γ)⁶⁰Co can also add greatly to the Compton continuum limiting the detection of other elements such as silicon, nickel, iodine, several rare earths and even zinc.



NAA and Resonance Integrals



Epithermal NAA Nuclear Reaction I_{ν}/σ_{ν}^{0}

⁵⁹Co(n,γ)^{60m}Co ¹⁸⁶W(n,γ)¹⁸⁷W ⁷⁵As(n,γ)⁷⁶As $^{109}Ag(n,\gamma)^{110}Ag$ 115 ln(n, γ) 116m ln ⁸¹Br(n,γ)⁸²Br 127 l(n, γ) 128 l 121 Sb(n, γ) 122 Sb ⁶⁸Zn(n,γ)^{69m}Zn ¹²⁴Sn(n,γ)^{125m}Sn ²⁹Si(n,p)²⁹Al

1.91 12.80 13.56 15.38 16.33 18.52 23.71 33.90 43.06 61.54

TRIGA Reactor





Compton Suppression

 Since early 1990's Compton suppression neutron activation analysis (CSNAA) has been effectively employed to quite dramatically lower detection limits for many elements

 Best exploited when the gamma ray of analytical interest is the only or major one that is involved in the beta decay process

Compton Suppression

- Many more institutions now have Compton suppression instrumentation in their laboratories
- Many more published papers on characterization, development and applications in NAA, environmental counting and fission product identification
- CSNAA can be judiciously used in biological specimens, but using ENAA can even further reduce backgrounds levels

Block Diagram of Compton Suppression System







Compton Suppression

 Compton suppression is ideal for radionuclides that emit single gamma-rays or gamma-rays that are not in coincidence with other photons in the decay scheme.

 Some examples include ¹³⁷Cs in environmental samples, ¹⁹⁸Au in neutron irradiated geological samples and ²⁰³Hg and ¹²⁸I in neutron irradiated biological samples.

Examples of Some Prime Radionuclides for CSNAA

⁶⁵Zn (1115 keV) ⁵²V (1434 keV) ⁵¹Ti (320 keV) ⁸⁰Br (618 keV) ²⁰³Hg (279 keV) ¹⁹⁸Au (411 keV) ⁵¹Cr (320 keV) ¹¹⁵Cd/¹¹⁵In (336 keV) ¹²⁸I (443 keV)

Decay Scheme of Single 279 keV Emitting Gamma Ray of the ²⁰³Hg Radionuclide

²⁰³Hg (46 day) Decay Scheme



Decay Scheme of Two Strongly Coincident Emitting 1368 and 2754 keV Gamma Rays of the ²⁴Na Radionuclide

²⁴Na (14.9 hr) Decay Scheme



Major Interferences Giving Rise to High Backgrounds or Spectral Interferences

- ²⁴Na 1368, 2754 keV
- ²⁸Al 1779 keV single emitting photon
- ³⁸Cl- 1642, 2167 keV
- ⁵⁶Mn 846, 1810, 2112 KeV
- ⁶⁰Co 1173, 1332 keV
- ⁴⁶Sc 889, 1120 keV
- ⁵⁹Fe 1098, 1291 keV
- ⁷⁵Se on ²⁰³Hg
- ${}^{239}U \rightarrow {}^{239}Np$ on ${}^{115}Cd/{}^{115}In$

Weakly Coincident Gamma Rays

- Some radionuclides that have two or more gamma rays in their decay can also benefit from CSNAA, if one of them has weaker coincidences with the remaining gamma ray(s).
- The radionuclides ⁷⁶As (559 keV) and
 ¹²² Sb (564 keV) are two such cases in point

NIST 1635 Coal Short-Lived NAA



NIST 2711 Soil



NIST 2711 Soil



Fully Automated Fast Pneumatic System for NAA



Fully Automated Fast Pneumatic System for NAA

- Analyze short-lived radionuclides
- Cyclic sample irradiations
- Minimize user interaction
 - reduce exposure
- Provide an efficient means of analyzing multiple samples

Considerations

Shielding

Detector Setup

 Outer Shield: Lead
 Inner Shield I: Cadmium
 Inner Shield II: Copper







Setup



Photon Attenuation

 Photon attenuation remains a problem that is constantly overlooked particularly when trying to correctly determine low energy gamma rays ordinary matrices.

• The problem is severely compounded when there samples have high-Z materials

Neutron Flux Monitoring

• Another area which needs attention is the monitoring of neutron fluence that a sample receives for short-lived NAA.

 While some reactors such as the 20 KW Canadian SLOWPOKE and the 30 KW Chinese Miniature Neutron Source reactor (MNSR) have very stable neutron fluxes that vary only 1-2 %, other reactors such as TRIGA have neutron fluences that vary ± 5-7%.

Rare-Earth Analysis

• Fission interferences

Spectral interferences

 It still remains surprising how many paper using NAA for rare-earth analysis do not take into account any of these interferences

Compton Suppression for Rare-Earth Analysis



Energy (keV)

New Directions

- In the recent past my group has been involved in the nuclear forensics and nuclear fuel cycle experiments.
- We have judiciously used NAA to produce fission products to test our low level Compton suppression system
- To produce surrogates for nuclear fuel cycle separation experiments conducted at national laboratories

Training and Teaching

- One are that is neglected is the pedagogical aspects of NAA.
- It has been 40 years that the last comprehensive NAA book has been written by Soete, Gijbels and Hoste (1972)
- While different aspects of NAA are covered in other nuclear and radiochemistry courses, there appears to be no course completely dedicated to NAA.
- Given the enormous advances that NAA has undergone, it behooves the community to write another book.

SUMMARY AND CONCLUSIONS

- Considering the costs of reactor resources, it is very advantageous to install both epithermal and Compton suppression systems to augment the array of elements that can be better determined in a variety of samples.
- While a cyclic system may only be beneficial for a few elements, the automation and control of irradiation, decay and counting times for short-lived NAA would greatly enhance quality assurance procedures.