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An evaluation of Compton suppression neutron activation analysis for determination of trace elements in some geological samples

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ABSTRACT

Compton suppressed neutron activation analysis has been used for a variety of applications, but never has a detailed discussion of its use in far more complex matrices, such as geological samples, been fully addressed. This investigation seeks to serve as a qualitative evaluation of Compton suppression neutron activation analysis (CSNAA) and to illustrate the benefits of using Compton suppression with thermal and epithermal neutrons for the analysis of several geological specimens.

1. Introduction

Compton suppression has been employed for the past 15 years primarily for neutron activation analysis of food (Anderson and Cunningham, 2008; Nyarko et al., 2008; Freitas et al., 2008), air filters (Biegalski and Landsberger, 1995) and soil (Landsberger and Wu, 1995). It has also been use for ^{137}Cs (Iskander et al., 2000) and naturally occurring radioactivity in environmental samples (Kapsimalis and Landsberger, 2008). There has been recent interest as to the applicability of Compton suppression in conjunction with neutron activation analysis for analysis of the far more complex matrices of geological samples (Steinnes, 2008). Issues that commonly arise when dealing with the neutron activation of geological samples are high dead times, especially for short-lived NAA, complex spectra, high Compton continuum and spectral interferences.

The high background of the spectra is caused by the abundance of radionuclides such as ^{24}Na ($t_{1/2}=15.02$ h), ^{28}Al ($t_{1/2}=2.24$ min), $_{38}$Cl ($t_{1/2}=37.2$ min), ^{56}Mn ($t_{1/2}=2.56$ h), ^{60}Co ($t_{1/2}=5.3$ y), ^{46}Sc($t_{1/2}=83.8$ d), ^{82}Br ($t_{1/2}=35.3$ h) and ^{59}Fe($t_{1/2}=44.5$ d). While these are the more common elements in geological samples that cause a high Compton continuum, there may be other constituents in samples of industrial waste or in ores that can give rise to high backgrounds as well. Specific spectral interferences are also an issue. Two examples are the 279 keV gamma ray of ^{75}Se interference on the 279 keV gamma ray ^{203}Hg or the 334 keV gamma ray of ^{239}Np (as a result of the ^{239}U → ^{239}Np+β reaction) on the 336 keV gamma ray of ^{115}Cd/^{115}In.

Compton suppression is best used when the gamma ray of analytical interest is the only or major gamma ray that is involved in the beta decay process. Prime examples of radionuclides that particularly benefit from Compton suppression are the following ones. For short-lived NAA we have: 1039 keV photopeak from ^{66}Cu; 388 keV photopeak from ^{88m}Sr, 165 keV peak from ^{139}Ba; 332 keV photopeak from ^{125m}Sn; 443 keV photopeak of ^{128}I; 320 keV photopeak of ^{51}Ti; and 1434 keV photopeak from ^{52}V. There is also the determination of 1279 photopeak of ^{29}Al for silicon determination through the through the ^{29}Si(n,p)^{29}Al reaction. For medium-lived NAA we have: 140 keV photopeak from ^{99}Mo; 411 keV photopeak from ^{198}Au; 439 keV photopeak from ^{69m}Zn; and 336 keV photopeak from ^{115}Cd/^{115}Id. For long-lived NAA we have: 1115 keV photopeak from ^{65}Zn; 279 keV photopeak from ^{203}Hg; and 514 keV photopeak from ^{85}Sr. The 810 keV photopeak from ^{58}Co also greatly benefits from Compton suppression. This is the analytical peak of nickel through the ^{58}Ni(n,p) ^{58}Co reaction. Of course, that is not to state that these are the only nuclides that can benefit from the use of Compton suppression in geological specimens. Some radionuclides that have two or more gamma rays in their decay can benefit from Compton suppression neutron activation analysis (CSNAA), assuming one has weaker coincidences with main photopeak of analytical interest. Two examples are the 559 keV peak of ^{76}As and the 564 keV peak of ^{122}Sb. Certain radionuclides that give rise to high backgrounds such as ^{24}Na, ^{56}Mn, ^{46}Sc, ^{37}Cl, ^{82}Br and ^{59}Fe, also have gamma rays that are strongly in coincidence with each other. The photopeaks are suppressed (and ultimately the Compton background as well), because the Compton system cannot resolve the difference between scattered and coincidence photons. Hence, photopeaks from both processes are suppressed. This means that there is even a further reduction of the Compton continuum, because the gamma rays responsible to these backgrounds are also significantly suppressed.

This work involved the investigation of the improvement of detection for several key trace elements in geological samples that

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are otherwise difficult, if not impossible, to detect using typical activation and counting techniques. In addition to the Compton suppression, the work compared the effects of using thermal versus epithermal neutrons for the reduction of several key interfering isotopes. Certain elements have relatively high integral or resonance cross-sections (I_γ/σ_γ). Many of the interfering elements such a scandium, sodium, chlorine and iron have low resonance cross-sections, whereas those with high ones are also elements of interest in NAA. These elements listed in Table 1.

2. Experimental

The Compton suppression system used was a high purity germanium detector, surrounded by Na(Tl) detectors. A complete description is given elsewhere (Landsberger, 1994). This investigation analyzed three geological standard reference materials, which included two coal samples (NIST 1635, NIST 1632a) and a soil sample (NIST 2711); each sample weighed roughly 0.5 g. The samples were irradiated in the University of Texas 1.1 MW TRIGA Mark II research reactor at 500 kW. The neutron flux densities were $\sim 2.1\times10^{12}\,cm^{-2}\,s^{-1}$ for thermal neutrons and $\sim 2.1\times10^{11}\,cm^{-2}\,s^{-1}$ for epithermal neutrons, respectively. For short-lived NAA, a pneumatic facility with electronic timers monitoring the exact irradiation and decay times were used. Typical irradiation, decay and counting conditions were 10 s, 15 min and 10 min, respectively for both thermal and epithermal NAA. Geometrical positions were chosen so that the dead-time was 5% or less to maximize the effect of the Compton suppression. Deadtimes above 10% begins to diminish the suppression due to random coincidences. For medium-lived NAA samples were placed in a rotary specimen rack (RSR). For the epithermal medium-lived NAA samples were placed in a rotating cadmium-lined tube with molybdenum flux wires. Typical irradiation, decay and counting conditions were 30 min, 2–3 d and 1 h, respectively. All these samples were then counted 2 weeks later for a period of 2 h for long-lived NAA.

Table 1
Some radionuclides that benefit from epithermal NAA.

Nuclear reaction	I_γ/σ_γ
$^{59}Co(n,\gamma)^{60m}Co$	1.91
$^{186}W(n,\gamma)^{187}W$	12.80
$^{75}As(n,\gamma)^{76}As$	13.56
$^{109}Ag(n,\gamma)^{110}Ag$	15.38
$^{115}In(n,\gamma)^{116m}In$	16.33
$^{81}Br(n,\gamma)^{82}Br$	18.52
$^{127}I(n,\gamma)^{128}I$	23.71
$^{121}Sb(n,\gamma)^{122}Sb$	33.90
$^{68}Zn(n,\gamma)^{69m}Zn$	43.06
$^{124}Sn(n,\gamma)^{125m}Sn$	61.54

3. Results

3.1. Short-lived

The first component of this investigation was to look at the entire gamma ray spectrum of the four methods (normal, Compton normal, epithermal and epithermal Compton). Fig. 1 shows the spectra obtained by the four methods and demonstrates the general reduction of background by using Compton suppression and epithermal neutrons for NIST Coal 1635. For all the spectra in this paper the following sequence is depicted: top spectrum, thermal normal, second to the top, epithermal normal, second from the bottom, Compton normal and bottom epithermal Compton. Fig. 2 depicts the absence of the 620 double escape peak of 1642 keV gamma ray of ^{38}Cl next to the 617 keV gamma ray of ^{80}Br. Often this can be a severe overlap making the analysis of the 617 keV gamma ray very difficult.

Several key peaks of the gamma ray spectra were then analyzed to demonstrate the effectiveness of the use of Compton suppression in conjunction with epithermal neutrons. Considering first the coal sample analyzed, a reduction in background and better counting statistics are evident by looking at the 320 keV photopeak of ^{51}Ti and the 388 keV photopeak of ^{87m}Sr. Fig. 3 shows the marked improvement of the background under these two photopeaks. Again, the Compton suppressed epithermal irradiations provided the best signal to background ratio of the

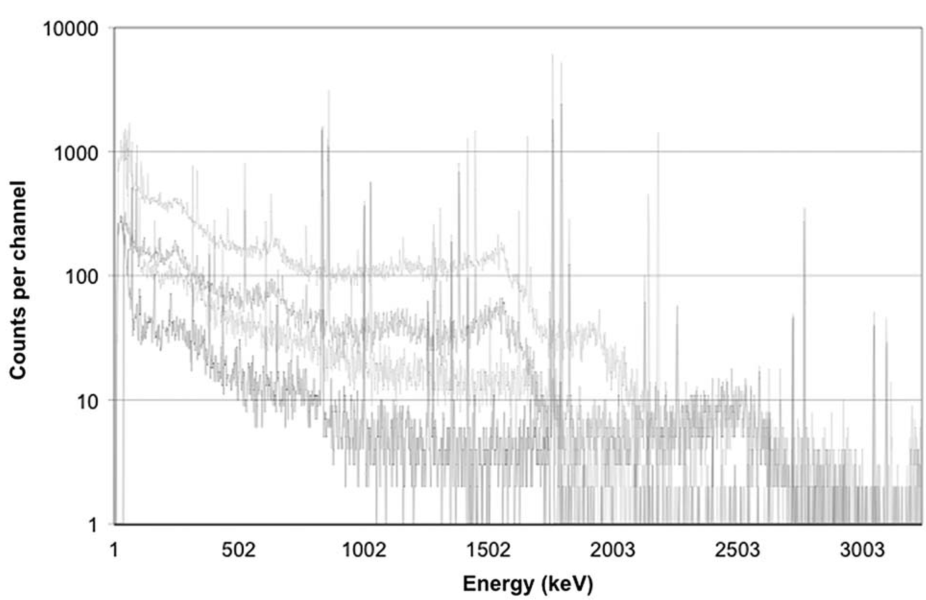

Fig. 1. General characteristics of the short-lived NAA of NIST Coal 1635 spectra. Top spectrum, thermal normal, second spectrum to the top, epithermal normal; second spectrum from the bottom, Compton normal; and bottom spectrum, epithermal Compton.