Advances in Instrumentation and Software for Radioanalytical Techniques

LOW-BACKGROUND GERMANIUM SPECTROMETRY - THE BOTTOM LINE THREE YEARS LATER

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Three years ago, state-of-the-art low-background germanium spectrometry was discussed, and speculations were advanced as to the origin of the remaining background. Some of those speculations have been shown to be incorrect. Contemporary lead shielding contains ~100 Bq/kg of 210Pb. Our 450-year-old lead was shown to contain <100 mBq/kg. A high purity electroformed copper Marinelli shield was placed around the detector with no effect on the background, which implied that the source is other than the 450-year-old shield. A new limit on the 210Pb in this old lead shield is <9 mBq/kg. Electroformed copper components were found to contain 226Ra and 228Th contaminations at levels of 3500 and 110 p.Bq/kg, respectively. High purity H₂SO₄, recrystallized CuSO₄, and a BaSO₄ scavenge in the electroforming bath have reduced these contaminations to <25 and 9 μBq/kg, respectively. In copper, cosmic ray induced nuclear reactions are now the dominant source of radioactivity. For example, 56Co can be readily measured after only a 24-hour exposure at sea level. A new germanium spectrometer containing 2150 grams of 87.44% enriched 76Ge has been fabricated to mitigate the effect of cosmogenic 68Ge in the background. Current background spectra are presented, and potential sources identified.

The premise of the following discussion is that there will occasionally be a need to make extremely selective and sensitive quantitative measurements of gamma-ray emitting radionuclides in various samples or materials. Specific examples are given in earlier contributions on this subject. Generally speaking, the limitation on sensitivity for measuring any specific gamma ray will be governed by the interfering radioactive background at the time of sample analysis, assuming signal enhancing features such as maximum practical sample size, detector efficiency, and counting time have been fully exploited. This background can be attributed to either the counting system or the sample itself. If the background introduced by the sample precludes measurement of the gamma ray of interest, steps to reduce the interfering background clearly must be taken or the measurement abandoned. These mitigation efforts could include coincidence/anticoincidence scenarios, electronic or pulse shape features, radiochemical separations, etc. This communication deals primarily with methods to reduce the system background to the lowest possible value so that it does not limit any particular measurement.

Two specific experiments which have been influential forces driving system background reduction efforts are double-beta decay experiments and detection of Cold Dark Matter interactions. Though the fundamental physics addressed in these experiments are extremely complex and exciting, they will not be discussed here, per se. However,
the results of our experiments to measure the double-beta decay of $^{76}$Ge will be used throughout this discussion as the example which has led to the best understanding of the sources of radioactive background and maximum effort to eliminate it. All steps regarding materials selection, electronic signal processing, and earth shielding from the effects of cosmic radiation are applicable and can be directly extrapolated to other counting systems and scenarios.

**Experimental**

The reference experiment is our measurement of the ordinary two-neutrino double-beta decay of $^{76}$Ge and efforts to measure the exotic zero-neutrino double-beta decay of the same isotope to the ground state of $^{76}$Se. "Ordinary" seems like a poor choice of words to describe the rarest event thus far discovered in nature. The half-life for this radioactive decay is on the order of $10^{21}$ years which makes it millions of times less probable than the longest half lives measured only a few years ago. The term "ordinary" refers here to the fact that there is nothing in our current understanding of the physics of radioactive decay which would preclude this event from happening. The zero-neutrino version of this decay mode would, however, require physics beyond the standard model of elementary particle physics and can be predicted by a number of different mechanisms. For example, if the neutrino has non-zero rest mass, this decay mode can be rationalized, which could explain the missing mass of the universe and significantly aid in the completion of a Grand Unified Theory. Neutrinoless double-beta decay of a $^{76}$Ge atom in a high resolution germanium diode gamma-ray spectrometer would be manifested as a monoenergetic event at the Q-value, 2038.56 keV, of the transition. Since the half-life for this hypothesized event is currently predicted to be on the order of $10^{25}$ years, the signal-to-noise ratio in the spectrometer will need to be more than simply extraordinary. The noise (background) will have to be virtually zero in order for the few events which may occur within the time frame of these experimenters' productive lives to be unequivocally identified as zero-neutrino double-beta decay of $^{76}$Ge.

The experimental arrangement has been extensively described earlier. A brief summary is, nevertheless, useful for understanding the current results. The facility is located nearly a mile underground (to minimize cosmic muons and induced background radioactivities) in the Homestake gold mine in Lead, South Dakota. An 11-ton passive lead shield (to minimize signals from the radioactivity in the mine environment) is lined inside with one ton of 450-year-old lead (to minimize $^{210}$ bremsstrahlung from the contemporary lead) and pressurized with the nitrogen gas boiling off from the cryogen (to minimize radon daughter activity). The current spectrometer system consists of a 2150 g hyperpure germanium diode composed of 87.44% isotopically enriched $^{76}$Ge (to maximize $^{76}$Ge sample size) contained in a cryostat assembly consisting of selected and/or fabricated components (to minimize radioactivity in the system components) which have been stored underground except during assembly and transported on the surface of the Earth rather than by air (to minimize cosmic neutron activation of the components).

**Results**

Modern lead typically contains a few hundred Bq/kg of $^{210}$Pb, and the bremsstrahlung from the $^{210}$Bi daughter beta particle can comprise the majority of the background up to ~850 keV in a reasonably good low-background system. Sources of old lead, i.e. ballast or freight from sunken Spanish, Dutch, German, or Roman ships, old