16th International Workshop on Low Temperature Detectors
20 24 July 2015

Simultaneous Measurement of MeV-Alpha Decays and keV-Beta Decays Inside a Microcalorimeter: Determining the Chemical Age of Trace-Level Plutonium Samples

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Determining the age of nuclear material collected by facility inspectors or through environmental sampling provides information about the material production history and is important for achieving safeguards and forensics goals. The 241Pu to 241Am decay chain is one method for measuring the time since chemical purification. Conventional approaches using mass-spectrometry nominally require radiochemical elemental separations, and often combine measurements from multiple methods (e.g. mass spectrometry, X-ray analysis, gamma-ray analysis). We report on a completely new approach, using a microcalorimeter detector, that can measure the chemical age via the 241Am/241Pu ratio, using a low-temperature transition-edge-sensor to measure the total reaction energy of decays from a sample embedded directly and completely inside an attached absorbing detector. Both isotopes are measured simultaneously in the same detector, with minimal chemical sample preparation requirements. The method is also non-consumptive, such that the material could be reacquired for additional analysis. The total nuclear reaction energy (Qalpha=5.6378 MeV) for the alpha-decay of 241Am is measured with very high fidelity and resolution. For the beta-decay of the 241Pu, the

measured energy has a continuum spectrum up to a maximum energy corresponding to the total reaction energy (Qbeta=20.78 keV). Since the sensors have a dynamic range ~6 MeV suitable for alpha-decays, the pulse heights for 241Pu decays are more than 2500-times small than those of 241Am. The very small pulse heights make quantification of the 241Pu challenging, and susceptible to both false positives (spurious events classified as beta decays) and false negatives (missed counts). There is also the potential for spectral distortion at low energy. We will report progress on efforts to diagnose and resolve these issues. (LA-UR-14-21402)