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Radiochronometry by Mass Spectrometry: Improving the Precision and Accuracy of Age-Dating for Nuclear Forensics

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The model-date of a nuclear material is a parameter established at the time it was last chemically purified. Assuming the material is homogeneous, this parameter is fixed and exact, but it may or may not be the same as the purification date. The decay of a radioactive parent to a radioactive or stable daughter is the basis of the radiochronometers that record this model-date. If upon purification, only the parent isotope is present in the material, then the model-date will be the same as the purification date. Otherwise, if any of the daughter isotope is present, then the model-date will be further in the past. Regardless, it is a fixed and characteristic signature of the material, and will not vary as long as the system remains closed, i.e., there is no post-purification fractionation of parent and daughter. Measurements of the parent-daughter pairs 234U-230Th, 235U-231Pa, 241Pu-241Am, 137Cs-137Ba and 90Sr-(90Y)-90Zr can be used to determine the model-dates of a variety of nuclear materials. All of these pairs are measured more precisely by mass spectrometric methods, because, for a given sample, more atoms can be measured by mass spectrometry than decays measured by radiometric methods. Also, some daughters, e.g. 137Ba and 90Zr, are stable isotopes and must be measured by means other than decay counting. The accuracy of the mass spectrometric analyses is determined by the reference materials used to calibrate the mass spectrometer and the spike materials used for isotope dilution analysis. The accuracy of the model-date is also dependent on the decay constants of the radionuclides. Improving the accuracy involves development of better-certified and new reference materials and spikes, and better measurements of relevant decay constants. Improving the measurement precision, in contrast, involves improvements to all aspects of the analyses to obtain greater signal/noise. These include improvements to the instrumental analytical methods (the way that samples are introduced or loaded, and the data collection schemes), the chemical purification methods (low blank, with high recovery and purity to eliminate isobaric interferences), and to the instruments themselves. State programs have recognized the importance of precise and accurate model-dates as a signature of a nuclear material, and efforts on all these fronts are being made internationally by national laboratories and institutions charged with developing standards and reference materials. Efforts within the United States include the production of new certified reference materials (U-Th and Cs-Ba radiochronometer standards) and spikes (229Th, 134Ba, 243Am, 236Np, 233U), and the development of guidance on the interpretation of radiometric data. Enhancement of radiochronometric methods and the development of tools needed to improve accuracy and precision are supported collaboratively by the U.S. Department of Homeland Security, U.S. Department of Justice Federal Bureau of Investigation, the U.S. Department of Energy at DOE National Laboratories and U.S. national metrology institutes.

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