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A Cooperation to Improve 231Pa/235U Age Dating Measurements of Uranium for Nuclear Forensics

Introduction
Increased requests for model age determination of nuclear material during nuclear forensic assessments have resulted in broad efforts by nuclear forensic laboratories to establish 231Pa/235U radiochronometry capabilities. Ingrowth of 231Pa from decay of 235U in chemically purified uranium provides a daughter/parent chronometry pair with which to age date uranium. The application of 231Pa/235U (as a second, independent chronometer) in combination with 230Th/234U can significantly expand confidence when determining model ages of uranium with unknown origin. However, developing a 231Pa/235U radiochronometry capability is challenged by a lack of available certified reference materials for quality control, as well as the short half-life of 233Pa, which is used as the 'spike'isotope for isotope dilution analysis of 231Pa. In an effort to address these challenges, the U.S. Department of Energy/National Nuclear Security Administration and the European Atomic Energy Community entered into a Joint Action Sheet for a 'Cooperation on 231Pa/235U Age Dating Measurements of Uranium for Nuclear Forensics.'An interlaboratory study between Lawrence Livermore National Laboratory (LLNL), Los Alamos National Laboratory (LANL), and the European Commission Joint Research Centre Karlsruhe (EC-JRC) was initiated in 2016.

Technical Work
The technical partnership between LANL, LLNL, and EC-JRC focused on interlaboratory 231Pa/235U model age determination in order to assess radiochronometry reproducibility and produce open-source 231Pa/235U model ages of a suite of uranium materials for the forensic community. An interlaboratory comparison was designed to jointly develop 231Pa/235U chronometry capabilities and measure model ages for three certified reference materials (CRMs): New Brunswick Laboratory CRM U100, CRM U630, and CRM 125-A. The scope of work included an exchange of procedures for the preparation and characterization of a 233Pa spike and radiochemical purification and mass spectrometry determination of 231Pa and 235U in bulk uranium materials. A newly produced 231Pa Nuclear Forensic Reference Material [1] was distributed between LANL, LLNL, and EC-JRC to support 233Pa spike calibration. Each laboratory used independent methods to determine 231Pa/235U radiochronometric model ages for separate units of CRM U100, CRM U630, and CRM 125-A [2, 3, 4]. The results obtained by the three laboratories are in very good agreement.

Summary
All laboratories have established 231Pa/235U capabilities, and results of the interlaboratory 231Pa/235U comparison will be presented here and interpreted in the context of nuclear forensics. The analytical results from this project support ongoing efforts to establish consensus 231Pa/235U ages for uranium CRMs for quality control to support nuclear forensic assessments for law enforcement and attribution. In addition to presenting analytical data, we will provide lessons learned and technical recommendations for the forensics community.

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[2] Eppich GR, Williams RW, Gaffney AM, Schorzman KC (2013) 235U-231Pa age dating of uranium materials for nuclear forensic investigations. Journal of Analytical Atomic Spectrometry 28, 666-674.

[3] Varga Z, Nicholl A, Hrnecek E, Wallenius M, Mayer K (2018) Measurement of the 231Pa/235U ratio for the age determination of uranium materials. Journal of Radioanalytical and Nuclear Chemistry 318, 1565-1571.

[4] Kayzar-Boggs TM, Treinen KC, Okubo A, Denton JS, Gaffney AM, Miller MM, Steiner RE, Wende AM, Williams RW (2020) An interlaboratory collaboration to determine consensus 231Pa/235U model ages of a uranium certified reference material for nuclear forensics. Journal of Radioanalytical and Nuclear Chemistry 323, 1189-1195.

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