

DEVELOPMENT OF A HIGH-PURITY Pa-233 SOURCE FOR THE YIELD  
DETERMINATION OF Pa-231 IN GASEOUS CENTRIFUGE COMPONENTS BY  
THERMAL IONIZATION MASS SPECTROMETRY

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Globally, gaseous centrifuge enrichment plants (GCEPs) represent a probable pathway in the development of fissionable nuclear materials. Compared to gaseous diffusion plants, GCEPs are relatively inexpensive to construct and operate. Furthermore, GCEPs can be concealed due to their compact size and low energy utilization / heat signature. While undeclared facilities represent the greatest proliferation risk, declared facilities are also a concern. Within declared plants, centrifuge cascades can be easily reconfigured to switch between low enriched U (LEU) and high enriched U (HEU) production.

The International Atomic Energy Agency (IAEA) would benefit from new forensic methods to determine historical enrichment operations within GCEPs. PNNL is presently working to exploit a new forensic signature that will enable the determination of centrifuge component historical use. The destructive method entails the analysis of enriched  $^{234}\text{U}$  and  $^{235}\text{U}$  daughter isotopes ( $^{230}\text{Th}$  ( $t_{1/2} = 7.54 \times 10^4$  y) and  $^{231}\text{Pa}$  ( $t_{1/2} = 3.28 \times 10^4$  y), respectively) that may be embedded in the corroded surface of the component. The quantity and ratio of these trace signatures should provide evidence of the average U enrichment run through the system, as well as the quantity of enriched U product produced. Since neither  $\text{ThF}_4$  nor  $\text{PaF}_5$  form volatile fluorides within the gaseous  $\text{UF}_6$  stream, it is expected that these molecules will deposit onto component surfaces.

Thermal ionization mass spectrometry (TIMS) will be utilized to measure  $^{231}\text{Pa}$  and  $^{230}\text{Th}$  by isotopic dilution analysis after a multi-column purification process is performed (still under development). While  $^{229}\text{Th}$  can be used as a readily available high-purity tracer for  $^{230}\text{Th}$ , a readily available and long-lived Pa tracer does not exist. With a half-life of 26.98 days,  $^{233}\text{Pa}$  (daughter of  $^{237}\text{Np}$ ) is the second longest lived isotope of Pa. In order to use  $^{233}\text{Pa}$  as a  $^{231}\text{Pa}$  tracer for TIMS, it is estimated that several pg will be required to add to the sample up front. Due to the great disparity between  $^{237}\text{Np}$  and  $^{233}\text{Pa}$  specific activities, the mass ratio at secular equilibrium is  $\sim 3 \times 10^7$ . In order to deliver the prepared samples to the TIMS clean room, the mass of  $^{237}\text{Np}$  contamination in the samples must be  $< 100$  pg. In other words, the  $^{237}\text{Np}$  decontamination factor from the  $^{233}\text{Pa}$  spike preparation is targeted to be  $\geq 3 \times 10^6$ . We will describe the development and optimization of the Pa/Np separation method development along with a description of the semi-automated fluidic workstation that is being assembled to perform this separation for use in the future.