Contribution ID: **116** Type: **Poster**

On demand production of a Pa-233 tracer

Uranium radiochronometry is of great interest to the nuclear forensics community [1–3]. The $^{235}U/^{231}Pa$ isotope pair can be used to calculate the separation age of samples containing enriched ^{235}U; a comparison with another radiochronometric pair, $^{234}U/^{230}Th$, can provide additional information about the processing history of the material. Accurate dating using the $^{235}U/^{231}Pa$ nuclear chronometer typically requires access to the short-lived radiotracer ^{233}Pa [$t_{1/2}$ = 26.98(2) d] [4], which is used to quantify the longlived ^{231}Pa ($t_{1/2}$ = 32.8 ky) by methods such as isotope dilution mass spectrometry [5, 6]. The short lived ^{233}Pa is ideal for decay counting primary standardization using coincidence counting and liquid scintillation counting (LSC) methods.

At the present time, access to ^{233}Pa is limited to the handful of laboratories around the world that possess significant quantities of ^{237}Np ($t_{1/2}$ = 2.14 My). As a decay product of ^{237}Np, ^{233}Pa forms continuously in situ, and can be isolated by complex chemical processing of these neptunium sources [5]. However, due to the long half-life of ^{237}Np, significant quantities of ^{233}Pa can only be generated after an interval of months from the previous source reprocessing, limiting its access upon immediate requisition. If the $^{235}U/^{231}Pa$ dating method is to be applied as a forensic tool in emergency response scenarios, access to ^{233}Pa must be improved significantly.

The deliberate generation of ^{233}Pa by bombarding natural thorium oxide with thermal neutrons was first reported more than 60 years ago [7]; but to our knowledge, this production route is not in use today. Although the reported post-irradiation processing is not appropriate for routine production due to the complex, multi-step chemical treatments and use of hazardous reagents including fluorine and hydrofluoric acid, the production physics of this process is near-ideal. Natural thorium is monoisotopic (100% ^{232}Th), and thus the sole product of its thermal neutron activation is the short-lived radioisotope ^{233}Th ($t_{1/2}$ = 22 min), which undergoes beta decay to the desired ^{233}Pa. The moderate cross-section for the $^{232}Th(n, \gamma)^{233}Th$ nuclear transformation (σ = 7.4 b) indicates that arbitrary (MBq) quantities of ^{233}Pa can be generated using this production route even in low- and medium-flux nuclear research reactors. A re-investigation of the separation of ^{233}Pa from neutron irradiated thorium to identify a methodology that is simple, rapid, and reproducible is therefore in order. The ideal approach would also avoid the use of reagents that require special handling, such as HF-assisted separations described elsewhere [8–10].

As one of the world's leading producers and processors of uranium, Canada is interested in expanding its nuclear forensics capabilities for characterizing samples that are found outside of regulatory control. Towards this goal, the objectives of this work were two-fold: first, to investigate the feasibility of "on-demand"production of ^{233}Pa from thorium at the McMaster Nuclear Reactor, the nation's most powerful research reactor; and second, to conduct standardization measurements at the National Research Council of Canada. The resulting certified reference materials could be used to calibrate secondary radioisotope quantification instrumentation.

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Track Classification: MORC: Nuclear forensics