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Calculation of Transactinide Homolog Isotope Production Reactions Possible with the Center for Accelerator Mass Spectrometry (CAMS) at Lawrence Livermore National Laboratory

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Introduction:

The LLNL heavy element group has been investigating the chemical properties of the heaviest elements over the past several years [1-5]. The properties of the transactinides (elements with $Z > 103$) are often unknown due to their low production rates and short half-lives, which require lengthy cyclotron irradiations in order to make enough atoms for statistically significant evaluations of their chemistry. In addition, automated chemical methods are often required to perform consistent and rapid chemical separations on the order of minutes for the duration of the experiment, which can last from weeks to months. Separation methods can include extraction chromatography, liquid-liquid extraction, or gas-phase chromatography. Before a lengthy transactinide experiment can be performed at an accelerator, a large amount of preparatory work must be done both to ensure the successful application of the chosen chemical system to the transactinide chemistry problem being addressed, and to evaluate the behavior of the lighter elemental homologs in the same chemical system. Since transactinide chemistry is literally performed on one single atom, its chemical properties cannot be determined from bulk chemical matrices, but instead must be inferred from the behavior of the lighter elements that occur in its chemical group and in those of its neighboring elements. By first studying the lighter group homologs in a particular chemical system, when the same system is applied to the transactinide element under investigation, its decay properties can be directly compared to those of the homologues, thereby allowing an inference of its own chemistry.

The Center for Accelerator Mass Spectrometry (CAMS) at Lawrence Livermore National Laboratory (LLNL) includes a 1 MV Tandem accelerator, capable of accelerating light ions such as protons to energies of roughly 15 MeV. By using the CAMS beamline, tracers of transactinide homolog elements can be produced both for development of chemical systems and for evaluation of homolog chemical properties. CAMS also offers an environment for testing these systems "online" by incorporating automated chemical systems into the beamline so that tracers can be created, transported, and chemically separated all on the shorter timescales required for transactinide experiments. Even though CAMS is limited in the types and energies of ions they can accelerate, there are still a wide variety of reactions that can be performed there with commercially available target materials. The half-lives of these isotopes vary over a range that could be used for both online chemistry (where shorter half-lives are required) and benchtop tracer studies (where longer lived isotopes are preferred.)

In this document, we present a summary of tracer production reactions that could be performed at CAMS, specifically for online, automated chemical studies. They are from chemical groups four through seven, 13, and 14, which would be appropriate for studies of elements 104-107, 113, and 114. Reactions were selected that had a) commercially available target material, b) half-lives long enough for transport from a target chamber to an automated chemistry system, and c) cross-sections at CAMS available projectile energies that were large enough to produce enough atoms to result in a statistically relevant signal after losses for transport and chemistry were considered. In addition, the resulting product atoms had to decay with an observable gamma-ray using standard Ge gamma-ray detectors. The table includes calculations performed for both metal targets and their corresponding oxides.

Table 1. Isotopes capable of being produced at CAMS for heavy element homolog chemistry experiments. Half-lives were chosen that were appropriate for online chemical studies using chemical automation.

Product Nuclide	Half-life	Gamma energy (keV)	Gamma branch (%)	Reaction	Projectile energy (MeV, lab frame)	Cross section (mb)	Target	Target thickness (mg/cm^2) [6]	Elemental thickness (mm or atoms/cm^2)^{a,b}	Instantaneous production rate (atoms/s/nA)	Refs.
Zr-89m	4.28 min	587.8 1507.3	89.50 6.10	^{89}Y(p,n)	10	370	Y metal Y_2O_3	0.026 0.02	6.00E-05 1.07E17	2.47E02	7-12
Nb-90m	18.8 sec	122.4	64	^{90}Zr(p,n)	10	460	Zr metal ZrO_2	0.025 0.018	3.80E-05 8.90E16	2.55E02	10, 13-15
Mo-91m	65 sec	652.9 1208.1 1508	48.2 18.7 24.3	^{92}Mo(p,pn)	15	12	Mo metal MoO_2	0.035 0.026	3.40E-05 1.26E17	9.43	12, 13, 15, 16-20
Tc-92	4.23 min	148 329.3 773 1509.6	71 79.9 100 100	^{92}Mo(p,n)	10	520	Mo metal MoO_2	0.023 0.017	2.30E-05 8.25E16	2.68E02	21-23
Hf-179m	18.7 sec	214.3	94	^{178}Hf(d,p)	13	250	Hf metal HfO_2	0.023 0.015	1.70E-05 4.30E16	6.71E01	24-26
Ta-178g	9.3 min	93.1 1340.8 1350.6	6.60 1.00 1.20	^{178}Hf(p,n)	11	70	Hf metal HfO_2	0.009 0.006	7.00E-06 1.73E16	7.56	11, 21, 27-29
W-185m	1.67 min	65.9 131.6 173.7	5.80 4.30 3.30	^{184}W(d,p) ^{186}W(α,αn) ^{184}W(^{3}He,2p)	13 38 28	10 20 25	W metal WO_3	0.021 0.012	1.10E-05 3.11E16	1.94	25-28, 30
Re-180	2.44 min	103.5 825.4 902.8	22.20 9.90 90	^{180}W(p,n)	11	150	W metal WO_3	0.009 0.005	5.00E-06 1.34E16	1.25E01	11, 21, 27, 31, 32

Tl-206g	4.20 min	216.4	74	^{205}Tl(d,p)	19	120	Tl metal	0.023	1.90E-05	3.14E01	25, 26, 28
		265.7	86				Tl_2O_3	0.016	4.20E16		
		453.3	93								
		686.5	90								
		1021.5	69								
Pb-201m	61 sec	629.1	54	^{199}Hg(α,2n)	24	300	Hg metal	0.062	n/a (liquid)	2.51E02	15, 27, 30, 33
							HgO	0.048	1.34E17		

[a] For metal targets, the elemental thickness is equivalent to the thickness of the metal foil given in mm. For oxides, the thickness is the number of target atoms of the element of interest per square centimeter.

[b] Assumes 100% enrichment of the oxide of the isotope of interest.

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