Preparation for online superheavy element homolog experiments using functionalized detector surfaces

A. S. Kirkland,^{1,2} R. Eichler,^{3,4} J. R. Garcia,^{1,2} I. W. Haynes,^{1,2} J. A. Mildon,^{1,2} P. Steinegger,^{3,5} E. E. Tereshatov,¹ G. Tiebel,^{3,5}

V. Zakusilova,¹ and C. M. Folden III^{1,2}

¹Cyclotron Institute, Texas A&M University, College Station, Texas 77843

²Department of Chemistry, Texas A&M University, College Station, Texas 77843

³Laboratory of Radiochemistry, Center for Nuclear Engineering and Sciences, Paul Scherrer Institute,

Forschungsstrasse 111, Villigen PSI, CH-5232, Switzerland

⁴Department of Chemistry, Biochemistry and Pharmaceutical Sciences, University of Bern,

Freiestrasse 3, Bern, CH-3012, Switzerland

⁵Laboratory of Inorganic Chemistry, Department of Chemistry and Applied Biosciences, ETH Zürich, Leopold-Ruzicka-Weg 4, Zürich, CH-8093, Switzerland

Superheavy elements (SHEs) characteristically have short half-lives and low production cross sections. This makes studying the chemistry of SHEs difficult, and improvements in experimental techniques are necessary in order to expand the understanding of SHE chemistry. A promising new technique for doing so is the use of self-assembled monolayers (SAMs) on the surface of gold-coated detectors. Recent work has shown that imidazole-based SAMs on the surface of gold chips can be used offline to selectively sorb rhodium (Z=45) and iridium (Z=77), homologs of meitnerium (Z=109), from solution [1]. Cyclotron-based experiments producing ¹⁷⁶Ir show promising results [2]. The current work describes a new system utilizing polonium (Z=84), an expected homolog of livermorium (Z=116).

Polonium was chosen due to the desirable decay properties of its isotopes and its relative availability. ²¹⁶Po ($t_{1/2} = 144$ ms, α -branching ratio: 100%) can be extracted in the gas-phase from a ²²⁸Th source utilizing a recoil transfer chamber (RTC) [3], which makes it viable to use for offline testing. Recent experiments by our group have also tested several nuclear reactions to determine the optimal conditions for producing suitable isotopes of Po in cyclotron-based experiments with the AGGIE gas-filled separator, and preliminary results are reported here.

These experiments were performed using the K150 cyclotron at Texas A&M University Cyclotron Institute. 40 Ar¹¹⁺ with an energy of 6.9 MeV/u was passed through a 15 µm Al window and various Al degraders, as well as a 2 µm Ti target backing. The targets used were 835 µg cm⁻² 160 Dy and 347 µg cm⁻² 162 Dy. The products were separated using the AGGIE gas-filled separator, which was set for a magnetic rigidity of 1.6 T m for both targets. The products were then focused onto two double sided silicon detectors (DSSDs).

The nuclear reactions performed include ¹⁶⁰Dy(⁴⁰Ar, xn)^{200-x}Po and ¹⁶²Dy(⁴⁰Ar, xn)^{202-x}Po. The latter is the most promising reaction, and an example spectrum, as well as the production rates as a function of the compound nucleus excitation energy are shown in Fig. 1. There are several isotopes produced, including ¹⁹⁷Po ($t_{1/2} = 84$ s, α -branching ratio: 44%), ¹⁹⁶Po ($t_{1/2} = 5.8$ s, α -branching ratio: 98%), and ^{195m}Po ($t_{1/2} = 1.92$ s, α -branching ratio: 98%).



FIG.1. (a) Preliminary example α -spectrum of three isotopes of Po using ¹⁶²Dy(⁴⁰Ar, xn)^{202-x}Po; "20+5 min" refers to 20 min of irradiation followed by a 5 min measurement without the ion beam on target to allow radionuclides to decay. (b) Preliminary excitation function for the same nuclear reaction; Rutherford counts refer to events collected on a pair of scattering detectors positioned ±30° respective to the ion beam axis, after it passes through any degraders and the target, scaling linearly with the luminosity (beam x target). Lines are drawn to guide the eye.

Since production of Po has been established, preparations have shifted towards the preparation of a wellorganized SAM. The chosen monolayer is 1,9-nonanedithiol (NDT; see Fig. 2(a)), which has a known thickness of 1.3 nm [4]. This is ideal for α -spectroscopy, since negligible amounts of energy will be lost in the monolayer. However, dithiol SAMs are prone to oxidation, which can complicate the self-assembly process and form disorganized multilayers [5]. Optimization of the deposition process is ongoing. Using spectroscopic ellipsometry, the measured layer thickness is 2.27 ± 0.031 nm (see Fig. 2(b)), which is indicative of a bilayer. Improvements to the deposition process are being considered, including changing the long alkyl chain to a more rigid phenyl-based backbone, and adding a reducing agent to the solution to prevent the oxidative coupling of the dithiol monolayer.

These experiments will continue with an online experiment to test the sorption of Po on the NDT SAM being planned for Fall 2024.



FIG.2. (a) Expected structure of 1,9-nonanedithiol (NDT) on an Au-coated glass wafer. (b) Spectroscopic ellipsometry data of NDT SAMs deposited from a 5 mM NDT solution in deoxygenated hexanes, corresponding to a NDT SAMs thickness of 2.27 ± 0.031 nm. This thickness is determined by measuring the phase difference (Δ) and amplitude difference (Ψ) of polarized light passing through the thin film, and a Cauchy model is used to determine the thickness.

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