Electrodeposition methods for an $^{225}Ac/^{213}Bi$ radionuclide generator with gold/silver nanolayers

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INTRODUCTION

Main Idea

Bismuth-213 is a short-lived ($t_{1/2}$=45.6 min) α-emitter of interest for targeted alpha-therapy (TAT).[1] Due to its short half-life, on-site $^{225}Ac/^{213}Bi$ radionuclide generators are required for research and clinical use. Current $^{225}Ac/^{213}Bi$ radionuclide generators use inorganic resins that fail at activities required for clinical use (> 100 mCi) due to the high linear energy transfer of α-particles.[2] This makes the development of novel generators with high radiolytic stability crucial to the success of ^{213}Bi-TAT radiotherapeutics.

Utilizing the Recoil Effect to Separate ^{225}Ac and ^{221}Fr Radionuclides

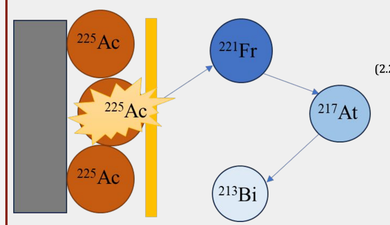
During ^{225}Ac alpha decay, ^{221}Fr receives ≈105 keV kinetic energy due to conservation of momentum which allows for the physical separation of ^{225}Ac and its daughter products. From this, we'll be able to develop a novel radionuclide generator system by electroplating ^{225}Ac onto Ni or Cu metal foils. Additionally, a thin Au/Ag coating is required enhance ^{225}Ac retention.

$$E_{recoil} = \left(\frac{m_\alpha}{m_{recoil}}\right) E_\alpha$$

Equations used to calculate ^{221}Fr recoil energy

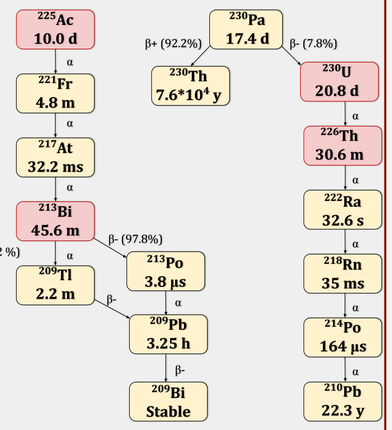
Reaction	Reduction Potential (V)
$Ag^+_{(aq)} + e^- \leftrightarrow Ag_{(s)}$	0.7996
$AuCl^-_{4\,(aq)} + 3e^- \leftrightarrow Au_{(s)} + 4Cl^-$	1.002

Reaction equations and Reduction potentials for Au and Ag



^{225}Ac plated on metal foil with thin gold nanolayer trapping parent and allowing escape of ^{221}Fr

Decay schemes for ^{225}Ac and ^{230}Pa



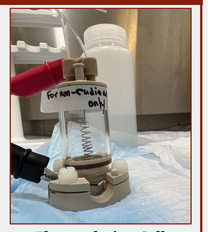
METHODS

Electroplating cell setup
- Platinum wire - anode
- Metal foil (Ni or Cu) - cathode
- 3 mL of solution with stirring (1 ml/min)
- Varied time, concentration, and voltage

Stopping Range of Ions in Matter (SRIM)[3]
- Monte Carlo program used to model charged particles traveling through different materials
- Estimated thickness of Au and Ag needed to maximize daughter escape

Scanning electron microscopy (SEM)
- Imaged foils to access coating uniformity and roughness
- Confirmed metal coating with EDS (not pictured)

Inductively coupled plasma-mass spectrometry (ICP-MS)
- Dissolved Ag/Ni foils in 6 M nitric acid with heat
- Determined total Ag addition

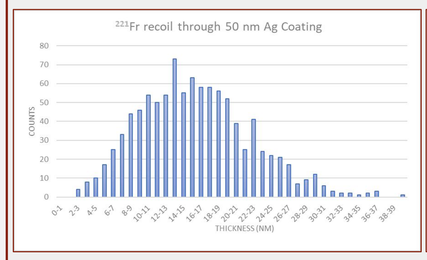


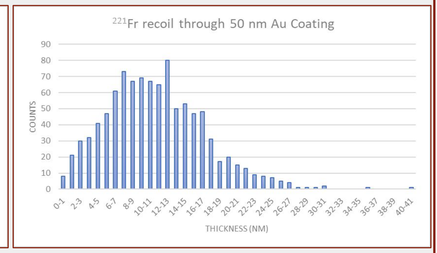
Electroplating Cell

Ni foils dissolved in Nitric Acid

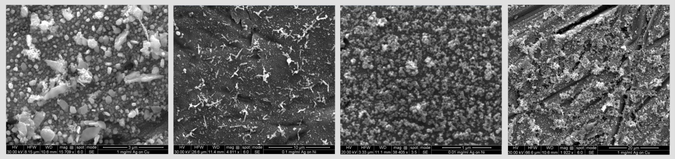
RESULTS

SRIM

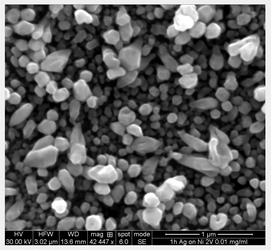




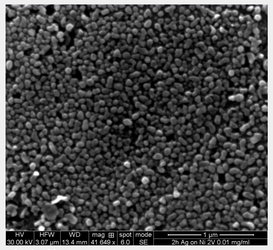
Electroplating & SEM

Silver Nitrate in 0.01 M Nitric Acid	1 mg/ml	0.1 mg/ml	0.01 mg/ml	0.001 mg/ml
SEM Images				

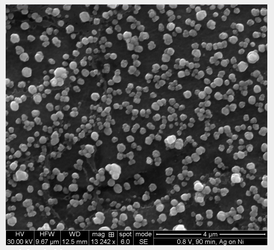
0.01 mg/ml Silver Nitrate in 0.1 M Nitric Acid



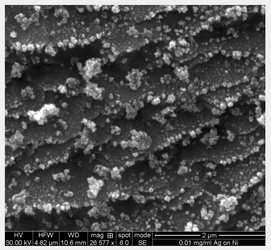
2 volt 60 minutes



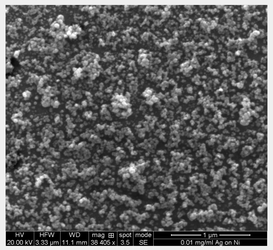
2 volt 120 minutes



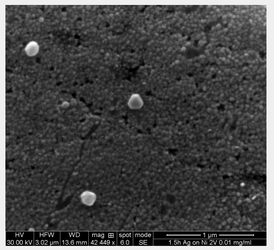
0.8 volt 90 min



Without mixing

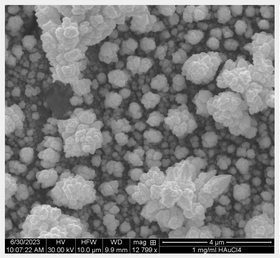


With mixing

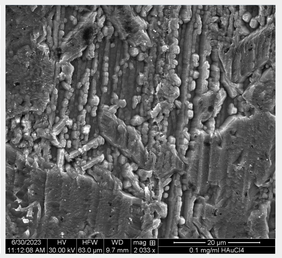


2 volt 90 min

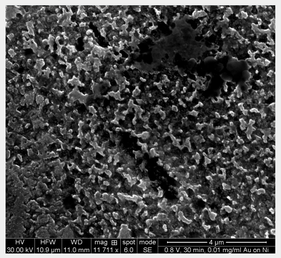
Varying Concentrations for Chloroauric Acid in 0.1 M Hydrochloric Acid



1 mg/ml $HAuCl_4$



0.1 mg/ml $HAuCl_4$



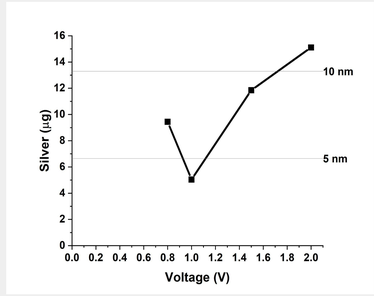
0.01 mg/ml $HAuCl_4$

RESULTS CONT.

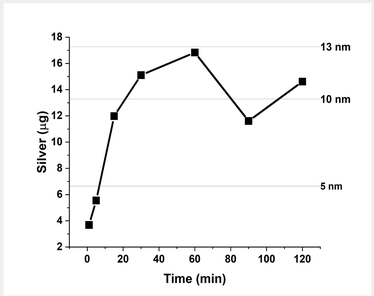
ICP-MS

- Based on the area of the electroplating cell, we were able to estimate coating thickness.
- Generally, the amount of silver deposited increased as voltage and time increased

Volts (V)	Time (min)	Metal (µg)	Thickness (nm)	Ag deposited (%)
2	1	3.7	2.8	19.33
2	5	5.6	4.2	29.15
2	15	12.0	9.0	62.89
2	30	15.1	8.9	62.21
2	60	16.8	3.8	26.47
2	90	11.6	7.1	49.56
2	120	14.6	11.4	79.30
0.8	30	9.4	12.7	88.35
1	30	5.0	8.7	60.94
1.5	30	11.9	11.0	76.70



30 min at varying voltage



2 volts at varying times

CONCLUSIONS & FURTHER WORK

Overall, our current research showed that thin nanometer coatings are possible which will be used in further development of the novel radionuclide generator system. With the current set-up, we were able to achieve a 10 nm coating for silver on Ni metal foils. Further research needs to be done on Au coatings, Cu metal foils, as well as electroplating consistency. These thin silver coatings will be investigated with ^{225}Ac and ^{230}U.

References

1. Birnbaum, Eva R., et al. "Actinides in medicine." The Heaviest Metals: Science and Technology of the Actinides and Beyond (2019): 445.
2. McDevitt, Michael R., et al. "An $^{225}Ac/^{213}Bi$ generator system for therapeutic clinical applications: construction and operation." Applied Radiation and Isotopes 50.5 (1999): 895-904.
3. Ziegler, James F., and Jochen P. Biersack. "The stopping and range of ions in matter." Treatise on Heavy-Ion Science: Volume 6: Astrophysics, Chemistry, and Condensed Matter. Boston, MA: Springer US, 1985. 93-129.

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HORIZON-BROADENING ISOTOPE PRODUCTION PIPELINE OPPORTUNITIES

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