Targetry Investigations of ^{186}Re Production via Proton Induced Reactions on Natural Osmium Disulfide and Tungsten Disulfide Targets

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Introduction

Radioisotopes play an important role in nuclear medicine and represent powerful tools for imaging and therapy. With the extensive use of ^{99m}Tc-based imaging agents, therapeutic rhenium analogues are highly desirable. Rhenium-186 emits therapeutic β^- particles with an endpoint-energy of 1.07 MeV, allowing for a small, targeted tissue range of 3.6 mm. Additionally, its low abundance γ-ray emission of 137.2 keV (9.42 %) allows for *in vivo* tracking of a radiolabeled compounds and dosimetry calculations. With a longer half-life of 3.718 days, synthesis and shipment of Re-186 based radiopharmaceuticals is not limited. Rhenium-186 can be produced either in a reactor or in an accelerator. Currently, Re-186 is produced in a reactor *via* the ^{185}Re(n,γ) reaction resulting in low specific activity which makes its therapeutic application limited.[1] Production in an accelerator, such as the PETtrace at the University of Missouri Research Reactor (MURR), can theoretically provide a specific activity of 34,600 Ci.mmol^{-1} Re[2], which represents a 62 fold increase over reactor produced ^{186}Re.

The studies reported herein focused on the evaluation of accelerator-based reaction pathways to produce high specific activity (HSA) ^{186}Re. Those pathways include proton and deuteron bombardment of tungsten and osmium targets by the following reactions: ^{186}W(p,n)^{186}Re, ^{186}W(d,2n)^{186}Re, ^{189}Os(p,α)^{186}Re, and ^{192}Os(p,α3n)^{186}Re. Additional information on target design related to the determination and optimization of production rates, radionuclidic purity, and yield are presented.

Material and Methods

Osmium and tungsten metals are very hard and thus very brittle. Attempts at pressing the pure metal into aluminum backings resulted in chalky targets, which easily crumbled during handling. Osmium disulfide (OsS_2) and tungsten disulfide (WS_2) were identified to provide a softer, less brittle chemical form for targets.

OsS_2 and WS_2 targets were prepared using a unilateral press with a 13 mm diameter die to form pressed powder discs. A simple target holder design (FIG. 1) was implemented to provide a stabilizing platform for the pressed discs. The target material was sealed in place with epoxy using a thin aluminum foil pressed over the target face.

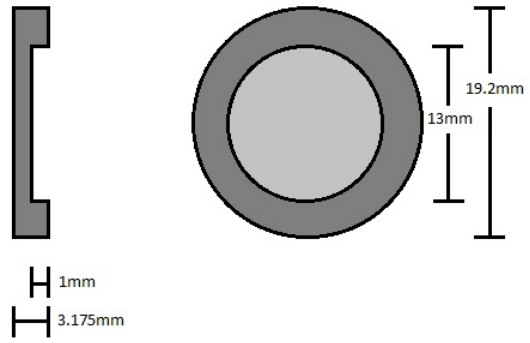

FIGURE 1. Aluminum backing design.

Initial irradiations of OsS_2 were performed using the 16 MeV GE PETtrace cyclotron at MURR. Irradiations were performed for 30–60 minutes with proton beam currents of 10–20 μA. Following irradiation, the OsS_2 targets were dissolved in NaOCl and the pH adjusted using NaOH. The resultant aqueous solution was mixed with methyl ethyl ketone (MEK), with the lipophilic perrhenate being extracted into the MEK layer and the osmium and iridium remaining in the aqueous layer. The MEK extracts were then passed through an acidic alumina column to remove any remaining osmium and iridium. Determination of rhenium and iridium activities was done by gamma spectroscopy on an HPGe detector (TABLE 1).

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Isotope	$t_{1/2}$	Gamma Energy (Intensity)
^{186}Ir	16.64 h	137 (23%), 297 (8.6%)
^{187}Ir	10.5 h	912 (4.3%)
^{188}Ir	1.72 d	155 (30%), 1210 (6.9%)
^{189}Ir	13.2 d	245 (6%)
^{190}Ir	11.8 d	187 (52%)
^{186}Re	3.718 d	137 (9.47%)
^{188}Re	17.004 h	155 (15.61%)
^{189}Re	24 h	216 (5.5%)

TABLE 1. Identified iridium and rhenium isotopes with their half-lives and utilized gamma emissions.

Preliminary irradiations on WS_2 targets were performed at MURR with the beam degraded to 14 MeV with a proton beam current of 10 µA for 60 minutes. After irradiation, WS_2 was dissolved using 30% H_2O_2 with gentle heating and counted on an HPGe detector to determine the radio-nuclides produced.

Results and Conclusion

Thin $^{nat}OsS_2$ targets were produced, irradiated at 16 MeV for 10 µAh, and analyzed for radiorhenium. Under these irradiation conditions, rhenium isotopes were produced in nanocurie quantities while iridium isotopes were produced in microcurie quantities (TABLE 2). Future studies with higher proton energies are planned to increase the production of rhenium and decrease the production of iridium. After optimizing irradiation conditions, enriched ^{189}Os will be used for irradiations to reduce the production of unwanted radionuclides.

Isotope	Activity (All aliquots)
^{186}Ir	3.00 µCi
^{187}Ir	143. µCi
^{188}Ir	10.0 µCi
^{189}Ir	9.00 µCi
^{190}Ir	1.34 µCi
^{186}Re	4.99 nCi
^{188}Re	4.19 nCi
^{189}Re	6.58 nCi

TABLE 2. Activities of produced iridium and rhenium isotopes at end of bombardment. 46 mg OsS_2 target, 16 MeV protons, 10 µAh.

A liquid-liquid extraction method separated the bulk of the rhenium from the iridium. The majority of the rhenium produced was recovered in the first organic aliquot with little iridium observed while the majority of the iridium and osmium was retained in the first aqueous aliquot.

Target production with WS_2 was successful. A thin target of $^{nat}WS_2$ was produced and irradiated at 14 MeV for 10 µAh. Under these irradiation conditions, several rhenium isotopes were produced in microcurie quantities. Target parameters to maximize ^{186}Re production remain to be determined before enriched ^{186}W targets are used for irradiations to reduce the production of unwanted radionuclides.

In conclusion, the potential production routes for accelerator-produced high specific activity ^{186}Re are being evaluated. Cyclotron-based irradiations of $^{nat}OsS_2$ targets established the feasibility of producing rhenium *via* the ^{nat}Os(p,αxn)Re reaction. Current results indicate higher proton energies are necessary to reduce the production of unwanted iridium isotopes while increasing the production of rhenium isotopes. Preliminary irradiations were performed using the 50.5 MeV Scanditronix MC50 clinical cyclotron at the University of Washington to determine irradiation parameters for future higher energy irradiations (20–30 MeV). A rapid liquid-liquid extraction method isolated rhenium from the bulk of the iridium and osmium following irradiation. Preliminary studies indicate WS_2 may also provide a suitable target material to produce ^{186}Re *via* the (p,n) reaction pathway.

References

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Acknowledgments

The authors would like to thank the University of Missouri Research Reactor staff and the University of Washington Clinical Cyclotron staff for conducting the irradiations necessary for this research. We would like to acknowledge the support of the United States Department of Energy through the Office of Science, Nuclear Physics (DE-SC0007348) and trainee support from the National Science Foundation under IGERT award DGE-0965983 (M.D. Gott) for funding this work.

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