Thick target preparation and isolation of ^{186}Re from high current production via the ^{186}W(d,2n)^{186}Re reaction

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Introduction

Rhenium-186 has a half-life ($t_{1/2}$ = 3.72 days) and emission of both gamma and beta particles that make it very attractive for use as a theranostic agent in targeted radionuclide therapy. ^{186}Re can be readily prepared by the ^{185}Re(n,γ)^{186}Re reaction[1]. However, that reaction results in low specific activity, severely limiting the use of reactor produced ^{186}Re in radiopharmaceuticals. It has previously been shown that high specific activity ^{186}Re can be produced by cyclotron irradiations of ^{186}W with protons and deuterons[2,3]. In this investigation we evaluated the ^{186}W(d,2n)^{186}Re reaction using thick target irradiations at higher incident deuteron energies and beam currents than previously reported. We elected not to use copper or aluminum foils in the preparation of our ^{186}W targets due to their activation in the deuteron beam, so part of the investigation was an evaluation of an alternate method for preparing thick targets that withstand μA beam currents.

Materials and Methods

Irradiation of ^{186}W. Initial thick targets (~600-1100 mg) were prepared using 96.86% enriched ^{186}W by hydraulic pressing (6.9 MPa) of tungsten metal powder into an aluminum target support. Those thick targets were irradiated for 10 minutes at 10 μA with nominal extracted deuteron energies of 15, 17, 20, 22, and 24 MeV.

Isolation of ^{186}Re. Irradiated targets were dissolved with H_2O_2 and basified with $(NH_4)_2CO_3$ prior to separation using column(s) of ~100–300 mg Analig Tc-02 resin. Columns were washed with $(NH_4)_2CO_3$ and the rhenium was eluted with ~80˚C H_2O. Gamma-ray spectroscopy was performed to assess production yields, extraction yields, and radionuclidic byproducts.

Recycling target material. When tested on a natural abundance W target, recovery of the oxidized WO_4^- target material from the resin was found to proceed rapidly with the addition of 4M HCl in the form of hydrated WO_3. The excess water in the WO_3 was then removed by calcination at 800 ˚C for 4 hours. This material was found to undergo reduction to metallic W at elevated temperatures (~1550 °C) in a tube furnace under an inert atmosphere (Ar). Quantification of % reduction and composition analyses were accomplished with SEM, EDS, and XRD and were used to characterize and compare both the WO_3 and reduced W_{metal} products to a sample of commercially available material.

Structural enhancement by surface annealing. In some experiments ~1 g WO_3 pellets were prepared from W_{metal} that had been chemically treated to simulate the target material recovery process described above. Following calcination, the WO_3 was allowed to cool to ambient temperature, pulverized with a mortar and pestle and then uniaxially pressed at 13.8 MPa into 13 mm pellets. Conversion of the WO_3 back to W_{metal} in pellet form was accomplished in a tube furnace under flowing Ar at 1550 °C for 8 hours. Material characterization and product composition analyses were conducted with SEM, EDS, and XRD spectroscopy.

Graphite-encased W targets. Irradiations were conducted at 20 μA with a nominal extracted deuteron energy of 17 MeV using thick targets (~750 mg) of *natural abundance tungsten* metal powder uniaxially pressed into an aluminum target support between layers of graphite powder (100 mg on top, 50 mg on the bottom) (FIG. 1). Targets were then dissolved as previously described and preliminary radiochemical isolation yields obtained by counting in a dose calibrator.

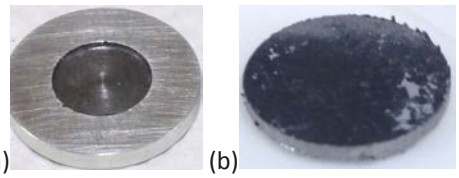

FIGURE 1. Structurally augmented graphite-tungsten sandwich pressed target pellets (a) in and (b) outside of the aluminum target support)

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Results and Conclusion

Although irradiations of W targets were possible at 10 μA currents, difficulties were encountered in maintaining the structural integrity of the full-thickness pressed target pellets under higher beam currents. This led to further investigation of the target design for irradiations conducted at higher beam currents. Comprehensive target material characterization via analysis by SEM, EDS, XRD, and Raman Spectroscopy allowed for a complete re-design of the target maximizing the structural integrity of the pressed target pellet without impacting production or isolation.

At the 10 μA current, target mass loss following irradiation of an enriched ^{186}W target was < 1 % and typical separation yields in excess of 70 % were observed. Saturated yields and percent of both ^{183}Re ($t_{½}$ = 70 days) and ^{184g}Re ($t_{½}$ = 35 days) relative to ^{186g}Re (decay corrected to EOB) are reported in TABLE 1 below. The reason for the anomalously low yield at 24 MeV is unknown, but might be explained by poor beam alignment and/or rhenium volatility during irradiation.

E_d [MeV]	^{186g}Re Y_{sat} [GBq/uA]	^{183}Re/^{186g}Re [%]	^{184g}Re/^{186g}Re [%]
15	1.47 ± 0.08	0.045 ± 0.003	0.38 ± 0.02
17	1.81 ± 0.09	0.131 ± 0.007	0.39 ± 0.02
20	1.96 ± 0.11	0.311 ± 0.021	0.45 ± 0.03
22	2.27 ± 0.12	0.448 ± 0.023	1.41 ± 0.07
24	1.61 ± 0.07	0.610 ± 0.042	4.43 ± 0.27

TABLE 1. Saturated yields and relative isotopic abundances in the enriched ^{186}W target at EOB.

Under these irradiation conditions, recovery yields of the W target material from the recycling process were found to be in excess of 90% with no discernable differences noted when compared to commercially available W_{metal} and WO_3 (FIG. 2).

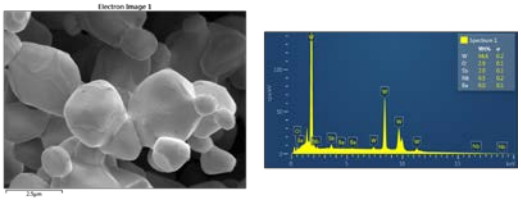

FIGURE 2. (Left panel) SEM image of target material after 1550 °C under Ar, and (right panel) Energy Dispersive X-Ray Spectroscopy analysis of material that was chemically processed

Conceptually, increasing the structural integrity of pressed WO_3 targets by high temperature heat treatment under an inert atmosphere is intriguing. However, the treated pellets lacked both density and structural stability resulting in disintegration upon manipulation (FIG. 3), despite the initially encouraging energy dispersive X-ray spectroscopy (EDS) determination that 94.9% percent of the WO_3 material in each pellet had been reduced to metallic W.

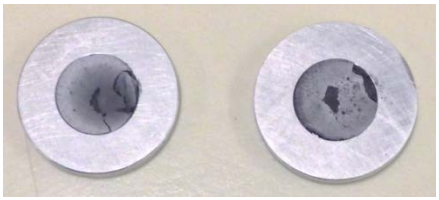

FIGURE 3. Targets containing 13 mm W_{metal} pellet made by reduction of uniaxially pressed WO_3. The cracks and indentations are indicative of the integrity issues that arise from incomplete pellet densification.

The use of powdered graphite as a target stabilizing agent provided successful irradiation of natural abundance W under conditions where non-stabilized targets failed (20 μA at 17 MeV for 10 minutes). Target mass loss following irradiation of a ^{nat}W target was < 1 % and a separation yield in excess of 97 % was obtained.

In conclusion, the theranostic radionuclide ^{186}Re was produced in thick targets via the ^{186}W(d,2n) reaction. It was found that pressed W metal could be used for beam currents of 10 μA or less. For deuteron irradiations at higher beam currents, a method involving pressing W metal between two layers of graphite provides increased target stability. Both target configurations allow high recovery of radioactivity from the W target material, and a solid phase extraction method allows good recovery of ^{186}Re. An effective approach to the recycling of enriched W has been developed using elevated temperature under an inert atmosphere. Further studies are underway with ^{186}W targets sandwiched by graphite to assess ^{186}Re production yields, levels of contaminant radiorhenium, power deposition, and enriched ^{186}W material requirements under escalated irradiation conditions (20 μA and 17 MeV for up to 2 hours).

References

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Acknowledgements

U.S. Dept. of Energy (DE-SC0007348) and NSERC of Canada Postdoctoral Fellowship (KG).

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