Abstract 009

A comparison of Nb, Pt, Ta, Ti, Zr, and ZrO_2-sputtered Havar foils for the high-power cyclotron production of reactive $[^{18}F]F^-$

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Introduction: Previous studies performed at the Edmonton PET Centre (EPC) have demonstrated that the use of Nb-sputtered Havar foils during $[^{18}F]F^-$ production via proton irradiation of $[^{18}O]H_2O$ decreases the radionuclidic and chemical impurities within the irradiated water[1]. Given the improved $[^{18}F]F^-$ reactivity, increased $[^{18}F]$FDG yield consistency, and decreased need for target rebuilding noted for Nb-sputtered Havar, these sputtered foils were adopted as the standard practice for $[^{18}F]F^-$ production at our facility in mid-2006. Following prolonged use of the Nb-sputtered foils however, degradation of the niobium film has been noted, with Havar impurities, FDG yield consistency and $[^{18}F]F^-$ reactivity returning over time to levels comparable with that of non-sputtered Havar.

Aim: The goal of this current work was to find a film that demonstrates increased longevity with regards to $[^{18}F]F^-$ reactivity when compared with niobium.

Methods: All film sputtering (Nb, Pt, Ta, Ti, Zr, and ZrO_2) was performed on 30 µm Havar at the University of Alberta's NanoFab micro and nanofabrication research facility (Edmonton, AB). Film thicknesses were verified through profilometer measurements and SEM micrographs.

To test the Havar impurity reducing properties of the sputtered foils (thicknesses = 250–450 nm), test irradiations were performed using 2.8–3.0 mL Barnstead 18MΩ-cm $^{nat}H_2O$. Multiple (N = 9–15) test irradiations (of 1,000 µAmin and 5,000 µAmin) were performed on all foils at 17.5 MeV using the EPC's TR 19/9 cyclotron to achieve total integrated currents of approximately 20,000–30,000 µAmin (weighted average currents of 69–81 µA). To ensure consistent irradiation conditions and complete sample transfer, both the ^{13}N saturated yield and the recovered $^{nat}H_2O$ mass were measured following all irradiations. Following ^{13}N decay, all water samples were assayed for radionuclidic impurities using an HPGe detector (dead time < 5%). Chemical analysis for extractable metals was also performed for a subset of the water samples via inductively coupled plasma mass spectroscopy (ICP-MS) at the Exova Lab (Edmonton, AB).

As tantalum was the only film which demonstrated Havar impurity-reducing properties comparable to niobium, the foil above was further irradiated to a total integrated current of 80,000 µAmin. Given the excellent continued performance noted via radionuclidic contaminant analysis, our next step was to install a new Ta-sputtered foil on our main production target for the purpose of testing both the $[^{18}F]F^-$ reactivity and evaluating the tantalum film's longevity performance. Prior to installation of the Ta-sputtered Havar on our production target, a series of five 1,000 µAmin (65 µA) $^{nat}H_2O$ test irradiations were performed on the existing (previously irradiated to ~980,000 µAmin) 400 nm Nb-sputtered Havar foil to establish a baseline to which the tantalum results could be compared. A new 900 nm Ta-sputtered Havar foil was installed and the produced $[^{18}F]F^-$ used for routine production of $[^{18}F]$FDG, $[^{18}F]$FAZA, and $[^{18}F]$FLT. Periodically (every 75,000–100,000 µAmin), a series of four test irradiations (1 @ 5,000 µAmin followed by 3 @ 1,000 µAmin) were carried out at 65 µA on $^{nat}H_2O$. All test irradiations were assayed for radionuclidic impurities.

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[1] Avila-Rodriguez, et al., *Appl. Radiat. Isot.* (2008) 66: 1775 [2] Wilson, et al., *Appl. Radiat. Isot*. (2008) 66: 565

Results: The following figure summarizes the Havar-associated radionuclidic impurities measured for the initial (approx. 20,000–30,000 µAmin) test irradiations, and the Ta-sputtered sputtered foil to 80,000 µAmin (“Ta (80k)”). With a clear dependence noted on the integrated current, the reported values are given as the average and standard deviation of the end-of-bombardment (EOB) radioactivity normalized to the integrated current for each irradiation. It is important to note that since the radionuclidic impurities showed a marked decrease for the first few irradiations on all new foils before reaching a relatively constant value, the first three 1,000 µAmin irradiations were omitted when producing the figure below. Evaluation of this figure reveals that tantalum is the only film which demonstrates radionuclidic impurity reducing characteristics similar to that of niobium. Based on strong correlations observed between the radionuclidic and ICP-MS measurements, we have concluded that trends noted in the radionuclidic impurities are reflective of trends in the ionic impurities.

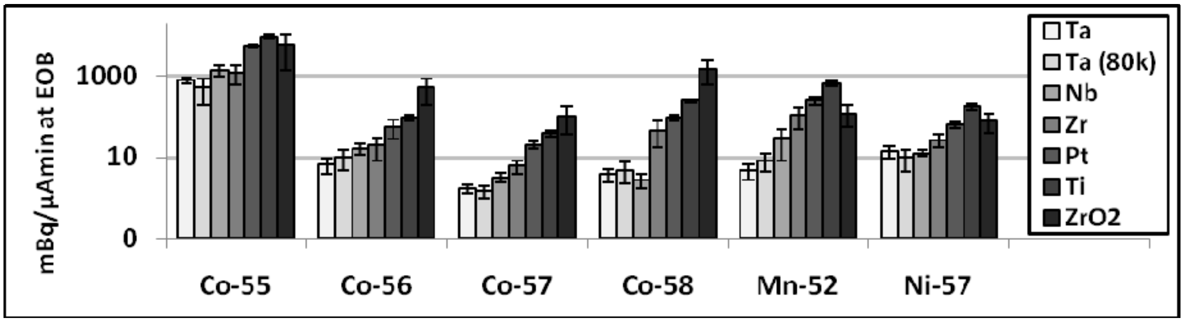

Table 1 summarizes the radionuclidic impurities (in units of mBq/µAmin at EOB) measured for the previously employed Nb-sputtered foil and the Ta-sputtered foil used on the production target. All values are reported as the average and standard deviation of the normalized activities. The integrated current (C) is reported as the total current on target prior to the test irradiations.

Table 1 **C [µAmin]**	**Nb** **979,307**	**Ta** **473,696**	**Ta** **1,0002,546**
Co-55	9748 ± 1621	37 ± 48	721 ± 238
Co-56	2038 ± 237	75 ± 27	171 ± 56
Co-57	807 ± 98	5 ± 1	13 ± 4
Co-58	9248 ± 1097	42 ± 6	120 ± 35
Mn-52	9035 ± 1476	98 ± 41	111 ± 48
Ni-57	2708 ± 394	18 ± 9	73 ± 18

Table 2 summarizes the [^{18}F]FDG decay-corrected (DC) yields and end-of-synthesis (EOS) activities (A) obtained on the EPC's GE TracerLab MX synthesis unit for all syntheses performed up to the reported integrated current. A comparison of the average [^{18}F]FDG DC yield (for comparable total integrated currents) demonstrates a 6.4 percent improvement (one-tailed t-test, $p = 0.0025$) with the Ta-sputtered foil when compared with the previously employed Nb-sputtered foil.

Table 2 **C [µAmin]**	**Nb** **936,802**	**Ta** **922,113**
N	38	35
Mean DC yield [%]	60.9 ± 11.7	67.3 ± 6.1
EOS $A_{average}$ [GBq]	123 ± 26	139 ± 19
EOS A_{max} [GBq]	171	184
EOS A_{min} [GBq]	64	109

Conclusions: Compared with our current Nb-sputtered Havar standard, the Ta-sputtered Havar demonstrates a significant reduction in the Havar-associated impurities following prolonged use up to ~1,000,000 µAmin. In addition to decreased Havar-associated impurities, we have also noted an improvement in the [^{18}F]FDG yields and yield consistency. Studies are currently underway to further evaluate this Ta-sputtered foil to a total integrated current of ~1,500,000 µAmin.

Acknowledgements: This project was supported by the University of Alberta’s MicroSystems Technology Research Initiative (MSTRI). The authors would like to thank Dr. Chris Backhouse and Ms. Eva Sant for their helpful discussions in film selection, and for performing the film sputtering.