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Assessing the performance and longevity of Nb, Pt, Ta, Ti, Zr, and ZrO₂-sputtered Havar foils for the high-power production of reactive [¹⁸F]F⁻ by proton irradiation of [¹⁸O]H₂O

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ABSTRACT

As water-soluble ionic contaminants, which arise following proton irradiation of [¹⁸O]H₂O have been associated with decreased [¹⁸F]FDG yields, the minimization of these contaminants is an asset in improving the [¹⁸F]F⁻ reactivity. To this end, we have previously demonstrated that the use of Nb-sputtered Havar foils results in decreased radionuclidic and chemical impurities in proton irradiated [¹⁸O]H₂O, improved [¹⁸F]FDG yields, and improved [¹⁸F]FDG yield consistency when compared with non-sputtered Havar. Resulting from the highly reactive chemical microenvironment within the target however, this niobium layer is observed to degrade over time. To find a material that displays increased longevity with regards to maintaining high [¹⁸F]F⁻ reactivity, this project extensively investigated and compared Havar foils sputtered with Nb, Pt, Ta, Ti, Zr and ZrO₂. Of the materials investigated, the results of this study suggest that Ta-sputtered Havar foil is the preferred choice. For similar integrated currents (~1,000,000 μA min), when comparing the Ta-sputtered Havar with Nb-sputtered Havar we observed: (i) greater than an order of magnitude decrease in radionuclidic impurities, (ii) a 6.4 percent increase ($p=0.0025$) in the average TracerLab MX [¹⁸F]FDG yield, and (iii) an overall improvement in the FDG yield consistency. Excellent performance of the Ta-sputtered foil was maintained throughout its ~1,500,000 μA min lifetime.

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1. Introduction

No carrier added [¹⁸F]F⁻ ($t_{1/2}=110$ min, $\beta^+=97\%$) is the most widely used radionuclide in positron emission tomography (PET) and is produced almost exclusively via proton irradiation of [¹⁸O]H₂O. Although this radionuclide is commonly employed for production of 2-[¹⁸F]fluoro-2-deoxy-D-glucose ([¹⁸F]FDG) via nucleophilic substitution, [¹⁸F]F⁻ is also used in the labeling of a variety of other PET radiotracers (e.g. [¹⁸F]FAZA, [¹⁸F]FMISO, [¹⁸F]FLT, [¹⁸F]FES). Furthermore, in light of the recent ^{99m}Tc isotope shortage, there is growing interest in using [¹⁸F]sodium fluoride as an alternative to ^{99m}Tc bone imaging agents (Hockley and Scott, 2010).

With the growing demand for [¹⁸F]F⁻, in large part due to the initial FDG-reimbursement approval in 1998 (Bietendorf, 2004),

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there have been continued efforts towards improving the target system design to allow for higher (> 1 kW) proton beam currents. One of the most important considerations required in the design of a high-power target is the selection of appropriate materials for both the target body and the entrance foil (Wilson et al., 2008). In addition to considering the mechanical strength and thermal performance of these materials, the effect of material selection on the [¹⁸F]F⁻ reactivity is also of importance.

Formation of ionic contaminants generated from beam interactions with the target body and entrance foil has been found to affect both the [¹⁸F]F⁻ reactivity and the labeling yield of radiopharmaceuticals (Kilbourn et al., 1985; Solin et al., 1988; O'Donnell et al., 2004). Over the last few years, target bodies have transitioned from materials such as titanium, stainless steel, and silver, to refractory metals such as niobium or tantalum. Selected for their chemical inertness, these refractory metals have lengthened the target maintenance intervals required for achieving high [¹⁸F]F⁻ reactivity.

Although the chemical inertness of niobium and tantalum are desirable properties for the target entrance foil, the weak mechanical

properties of these materials (tensile strengths of 585 and 650 MPa, respectively) make them unsuitable for [^{18}F]F $^-$ production under pressurized conditions. With its high tensile strength (1860 MPa), high melting point (1480 °C), and moderate thermal conductivity (14.7 W m $^{-1}$ K $^{-1}$ at 23 °C), Havar[®], a non-magnetic alloy (Co 42%, Cr 19.5%, Fe 19.3%, Ni 12.5%, W 2.6%, Mo 2.2%, Mn 1.7%, and C 0.2%) has grown to be the most popular choice as an entrance foil. One of the primary disadvantages to using Havar as an entrance foil however is that it gives rise to a wide variety of radionuclidic (Marengo et al., 2008; Avila-Rodriguez et al., 2008; Bowden et al., 2009) and chemical (Kilbourn et al., 1985; O'Donnell et al., 2004; Avila-Rodriguez et al., 2008) impurities in the irradiated water. While the ionic impurities resulting from the Havar foil may lead to decreased [^{18}F]F $^-$ reactivity, the presence of long-lived radionuclidic impurities also poses a possible concern for increased patient dose and the need for long-lived radioactive liquid waste and/or QMA cartridge/cassette storage.

Earlier work by our group at the Edmonton PET Centre has demonstrated that it is possible to decrease the Havar-associated contaminants in the irradiated water using Nb-sputtered Havar entrance foils (Avila-Rodriguez et al., 2008). This combination of the robust mechanical properties of Havar and the chemical inertness of niobium led to improved [^{18}F]F $^-$ reactivity, increased [^{18}F]FDG yield consistency, and decreased frequency of target rebuilding (Wilson et al., 2008). As a result, Nb-sputtered Havar was implemented as a standard component for [^{18}F]F $^-$ production at our facility in mid 2006. Following prolonged exposure to the proton beam however, the formation of a white film (presumably Nb $_2$ O $_5$) was noted on the Nb-sputtered surface (Wilson et al., 2008). Furthermore when changing the Nb-sputtered foils (due to decreases noted in the [^{18}F]FDG yields), we have noticed regions over the beam spot, which appear to be completely devoid of niobium. These observations led us to the conclusion that the niobium layer degrades over time due to the highly reactive chemical microenvironment at the foil/water interface. The goal of this current work was to find a film material that displays increased longevity with regards to the [^{18}F]F $^-$ reactivity.

2. Materials and methods

2.1. Film sputtering

As described by Sigmund (1987), sputtering is a standard method of film deposition. In this work, films of Nb, Pt, Ta, Ti, Zr, and ZrO $_2$ were sputtered onto 30 μm Havar (Hamilton Precision Metals Inc., Lancaster, PA, USA) via DC planar magnetron sputtering at the University of Alberta's micro and nanofabrication research facility ("Nanofab", Edmonton, AB). These materials were selected based on their chemical properties and ease of availability. For all film sputterings, an argon pressure of 7 mTorr and a power of 300 W were maintained. The voltages employed ranged from 325–450 V. Metallic sputtering targets (Kurt J. Lesker, at least 99.9% purity) were employed for all depositions, and the Havar foil was prepared as described previously (Wilson et al., 2008). The ZrO $_2$ film was achieved by reactive sputtering (Venkataraj et al., 2002)—a technique whereby O $_2$ gas is introduced during the sputtering process (in our setup, an O $_2$ flow rate of 3.5 sccm was employed). Film thicknesses (Table 1) were confirmed using a stylus-based measurement with an Alphastep 200 profilometer (Tencor Instruments).

2.2. SEM imaging

It has been suggested (Avila-Rodriguez et al., 2008) that the impurities, which arise in the water following the irradiation of a sputtered Havar foil, are a result of microporosity defects in the deposited film. In an attempt to understand the differences noted

Table 1

A summary of the irradiation conditions employed for the initial test irradiations.

Sputtered layer	Approximate thickness (nm)	N	Total current ($\mu\text{A min}$)	Weighted average current (μA)	^{13}N saturated yield (MBq/ μA)
Nb	400	12	28,001	81	1255 \pm 36
Pt	360	11	24,029	69	1068 \pm 131
Ta	350	13	32,505	78	1261 \pm 42
Ti	250	9	17,047	74	1150 \pm 80
Zr	375	15	31,001	79	1257 \pm 32
ZrO $_2$	450	13	29,000	80	1219 \pm 39

between the sputtering materials, the topographical structure of the sputtered films were examined via SEM imaging using a JAMP-9500F (JEOL) Field Emission Scanning Auger Microprobe at the Alberta Centre for Surface Engineering and Sciences (ACSES, Edmonton, AB). Because of the excellent strength of Havar (the property that makes it desirable here), obtaining clean fracture cross sections of the Havar (for purpose of imaging) was a rather difficult task. To this end, clean breaks were achieved (roughly half the time) by loading the sputtered Havar foil into a target (off-line from the cyclotron), and applying \sim 1000 psi Argon overpressure until the Havar foil ruptured. As this strategy was wasteful with regards to materials, when available, sputtered-aluminum foils were examined instead (since clean breaks were readily achieved). Due to the high radioactivity of the foils following irradiation, SEM images were only obtained on non-irradiated foils. In addition to providing information regarding the topographical structure of the sputtered films, the SEM images also served as a method for confirming the accuracy of the profilometer thickness measurements.

2.3. Irradiation conditions and target setup

For the purpose of selecting the optimal sputter coating, four sets of irradiations were performed in this study and all attempts were made to handle the $^{\text{nat}}\text{H}_2\text{O}$ test samples as consistently as possible. All irradiations referred to as "test irradiations" were performed using 17.5 MeV protons on 2.8–3.0 mL Barnstead 18 M Ω -cm $^{\text{nat}}\text{H}_2\text{O}$ with the Edmonton PET Centre's variable energy negative ion TR 19/9 cyclotron (Advanced Cyclotron Systems, Inc., Richmond, BC). Natural water was employed for these test irradiations due to the high cost associated with enriched [^{18}O]H $_2\text{O}$, the extensive number of test irradiations performed in this experiment ($N > 150$), and previous experiments, which demonstrated that there were no significant differences in impurity levels between enriched and natural abundance water (Avila-Rodriguez et al., 2008). In total, four sets of test irradiations were performed in this study:

- (1) All sputtered foils were irradiated multiple times to total integrated currents of approximately 20,000–30,000 $\mu\text{A min}$ (Table 1). These test irradiations consisted of multiple 1,000 $\mu\text{A min}$ (65 μA) irradiations, followed by one or more 5,000 $\mu\text{A min}$ (90 μA) irradiations, and finally several additional 1,000 $\mu\text{A min}$ (65 μA) irradiations.
- (2) From an analysis of the initial test irradiations, tantalum appeared to be the most promising candidate in reducing the radionuclidic impurities. As such, additional 1,000 $\mu\text{A min}$ (65 μA) and 5,000 $\mu\text{A min}$ (90 μA) test irradiations were performed to further evaluate the performance of the above noted Ta-sputtered Havar foil to a total integrated current of 80,000 $\mu\text{A min}$.

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