Supplemental Information

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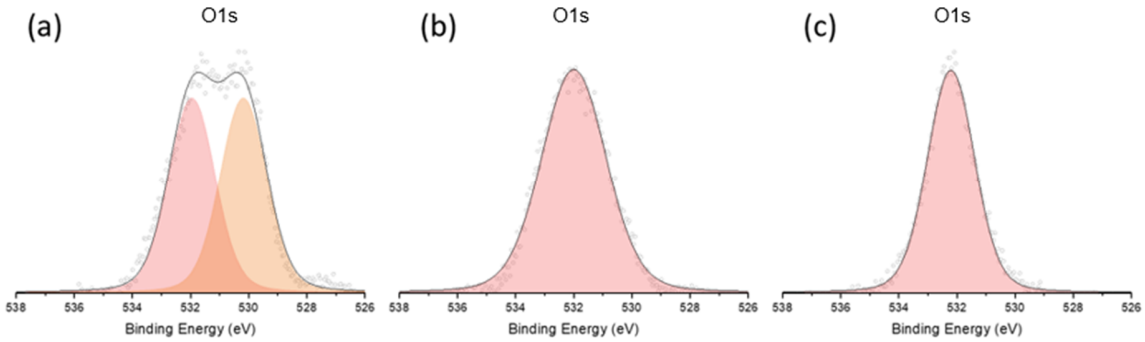

Figure S1. XPS O1s spectra for (a) pristine, (b) chemically etched, and (c) electropolished Ta foil.

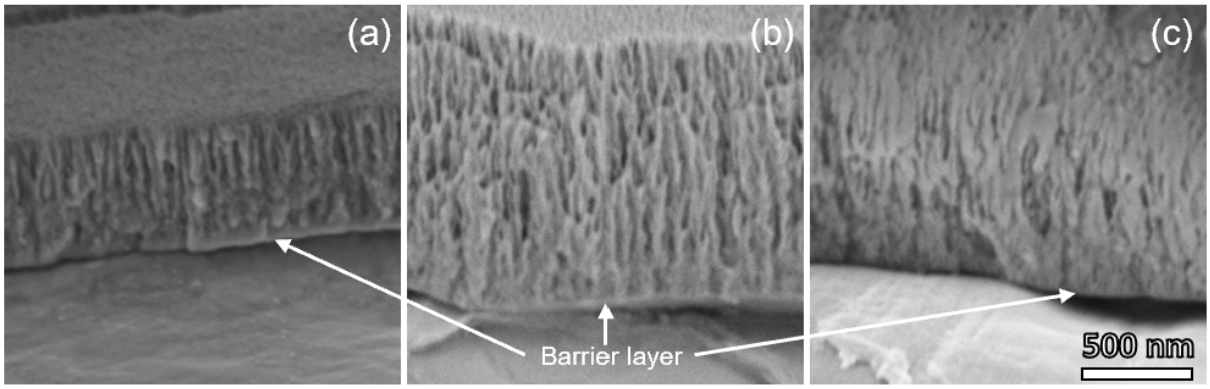

Figure S2. Cross-sectional SEM of anodic tantalum oxide films grown at 20V180°C on (a) electropolished, (b) chemically etched, and (c) pristine substrates, with the barrier layer indicated via arrows. The 500 nm scale bar is applicable to all figures.

Table S1. Quantitative oxidation-state fractions for XPS of pristine, chemically-etched, and electropolished samples.

Oxidation State	Ta^{5+}	Ta^{0}	Ta suboxide
Pristine	72.26%	24.44%	3.30%
Chemically Etched	79.28%	12.35%	8.37%
Electropolished	80.25%	9.97%	9.78%

Table S2. Pore sizes and wall thicknesses for films grown on chemically etched and electropolished substrates.

		Chemically Etched				Electropolished			
Voltage	**Temperature**	**Pore Size (nm)**	**Pore Density (pores/nm^2)**	**Pore Area (% of surface area)**	**Wall Thickness (nm)**	**Pore Size (nm)**	**Wall Thickness (nm)**	**Pore Density (pores/nm^2)**	**Pore Area (% of surface area)**
20V	170°C	17.9 +/- 2.14	0.14	54%	13.0 +/- 2.85	15.40 +/- 2.22	10.46 +/- 1.71	0.116	31%
	180°C	18.10 +/- 2.20	0.144	59%	11.31 +/- 2.43	21.25 +/- 2.48	6.86 +/- 1.40	0.108	25%
	190°C	15.84 +/- 1.73	0.148	64%	10.58 +/- 2.36	25.15 +/- 4.89	8.78 +/- 1.5	0.12	34%
30V	170°C	16.05 +/- 2.1	0.132	45%	10.69 +/- 2.47	16.80 +/- 1.89	10.7 +/- 1.52	0.112	28%
	180°C	14.54 +/- 2.02	0.136	49%	7.93 +/- 1.57	22.16 +/- 2.31	7.69 +/- 1.69	0.108	25%
	190°C	16.20 +/- 2.04	0.128	41%	7.24 +/- 1.36	29.71 +/- 3.42	7.41 +/- 1.55	0.132	45%
40V	170°C	15.81 +/- 1.97	0.12	34%	8.16 +/- 2.2	18.6 +/- 1.87	9.16 +/- 1.59	0.128	41%
	180°C	14.16 +/- 2.02	0.136	49%	8.85 +/- 1.27	22.31 +/- 2.71	7.04 +/- 1.93	0.132	45%
	190°C	17.07 +/- 2.4	0.156	75%	5.58 +/- 0.99	41.12 +/- 5.37	7.44 +/- 1.67	0.12	34%