# Supporting Information

"$CuNb_{1-x}Ta_xO_3$ ($x \leq 0.25$) Solid Solution: Impact of Ta(V) Substitution and Cu(I) Deficiency on its Structure, Photocatalytic, and Photoelectrochemical Properties"

Brandon Zoellner,[a] Sean Stuart,[b] Ching-Chang Chung,[c] Daniel B. Dougherty,[b] Jacob Jones,[c] and Paul A. Maggard*[,a]

*[a]Department of Chemistry, [b]Department of Physics, and [c]Department of Materials Science and Engineering, North Carolina State University, Raleigh, NC 27695-8204, United States*

**Table S1.** An example listing of the Miller (*hkl*) indices used for lattice parameter refinements of $CuNb_{1-x}Ta_xO_3$ in LATCON.

| Miller Indices (*hkl*) | Calculated 2θ (°) |
|---|---|
| 001 | 13.075 |
| 110 | 14.030 |
| 11-1 | 19.134 |
| 111 | 19.311 |
| 021 | 24.846 |
| 220 | 28.276 |
| -112 | 29.832 |
| 310 | 30.153 |
| -221 | 31.142 |
| 20-2 | 32.258 |
| 202 | 32.693 |
| 130 | 33.186 |
| 022 | 33.935 |
| 400 | 37.884 |
| 312 | 40.708 |
| 040 | 42.823 |
| 330 | 42.985 |
| -331 | 44.998 |
| 240 | 47.096 |
| -332 | 50.885 |
| -422 | 51.465 |
| 422 | 52.059 |
| 004 | 54.182 |
| -224 | 61.968 |
| 60-2 | 64.644 |
| -134 | 64.903 |
| 532 | 65.753 |

**Table S2.** Refined atomic positions, fractional occupancies, and $U_{iso}$ values for the atomic positions in $Cu_{1-x}NbO_3$.

| Atom | x | y | z | Site Occupancy | $U_{iso}$ |
|---|---|---|---|---|---|
| Cu 1 | 0.00000 | 0.1702(3) | 0.50000 | 0.983 | 0.0212(15) |
| Cu 2 | 0.7671(4) | 0.00000 | 0.2869(6) | 0.948 | 0.0217(16) |
| Nb 1 | 0.00000 | 0.20067(25) | 0.00000 | 1.000 | 0.0040(8) |
| Nb 2 | 0.25060(28) | 0.00000 | 0.2340(4) | 1.00 | 0.0185(11) |
| O 1 | 0.3640(12) | 0.1550(11) | 0.1037(17) | 1.00 | 0.016(3) |
| O 2 | 0.0764(10) | 0.8265(10) | 0.2557(16) | 1.00 | 0.012(4) |
| O 3 | 0.1237(13) | 0.00000 | 0.9601(22) | 1.00 | 0.010(6) |
| O 4 | 0.3404(14) | 0.00000 | 0.4939(22) | 1.00 | 0.005(5) |

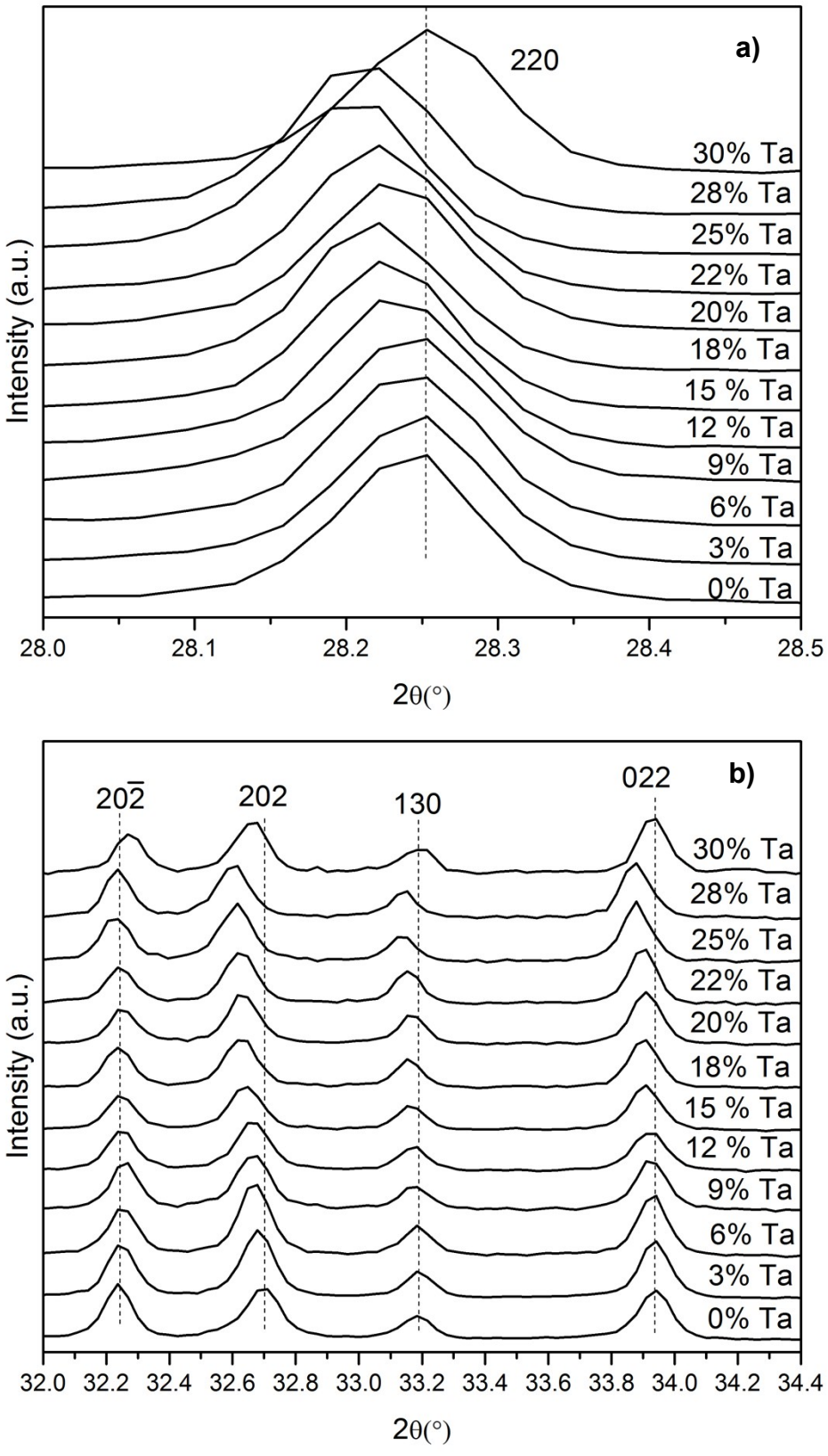


**Figure S1.** Powder X-ray diffraction of $CuNb_{1-x}Ta_xO_3$ with the 220 peak (a) and the 202, 130, and 022 peaks (b) shifting to lower 2θ with the increasing Ta(V) percentage.

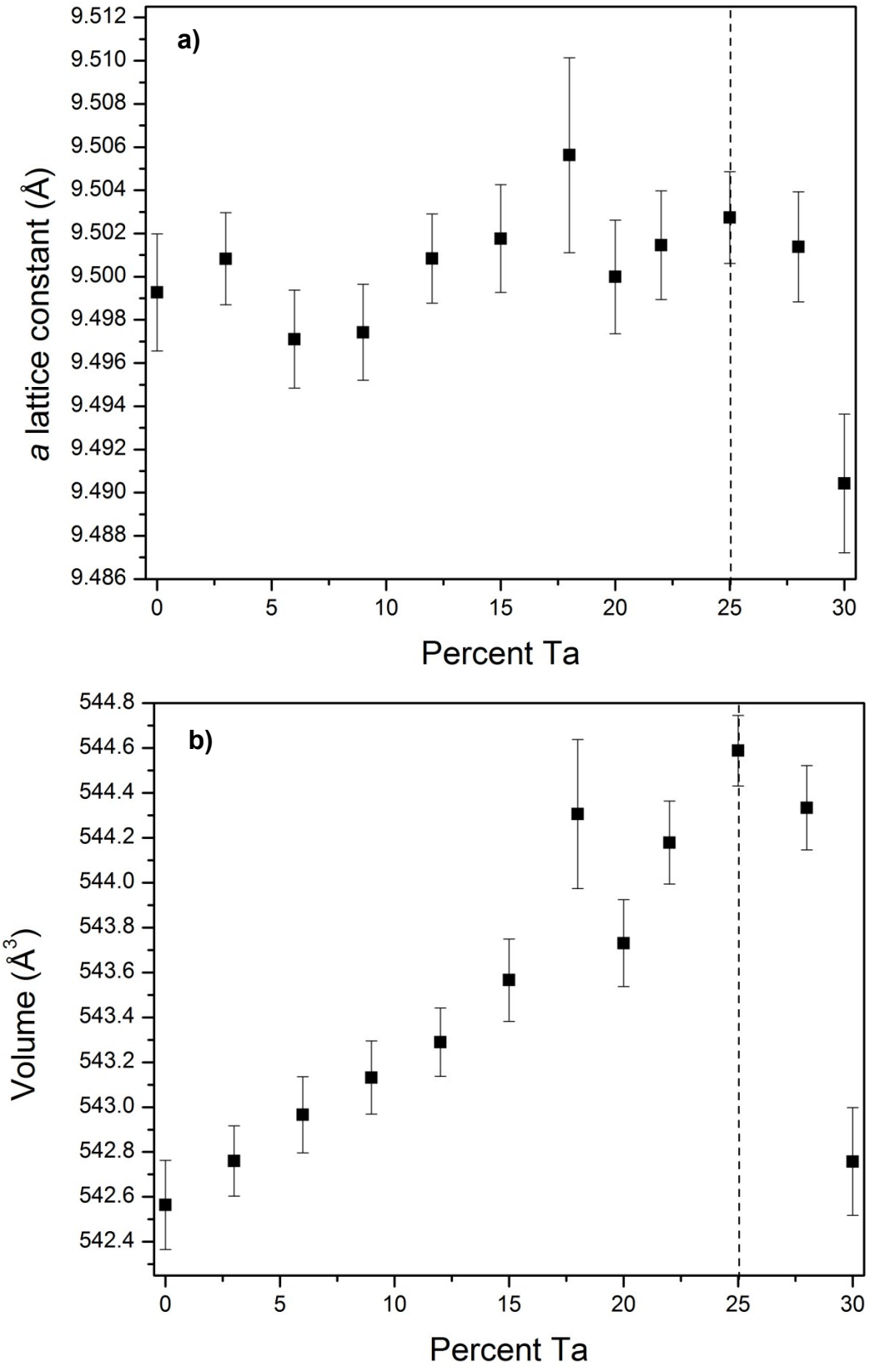


**Figure S2.** Refined lattice parameter *a* (a) and unit cell volume (b) of $CuNb_{1-x}Ta_xO_3$ ($0 \leq x \leq 0.25$).

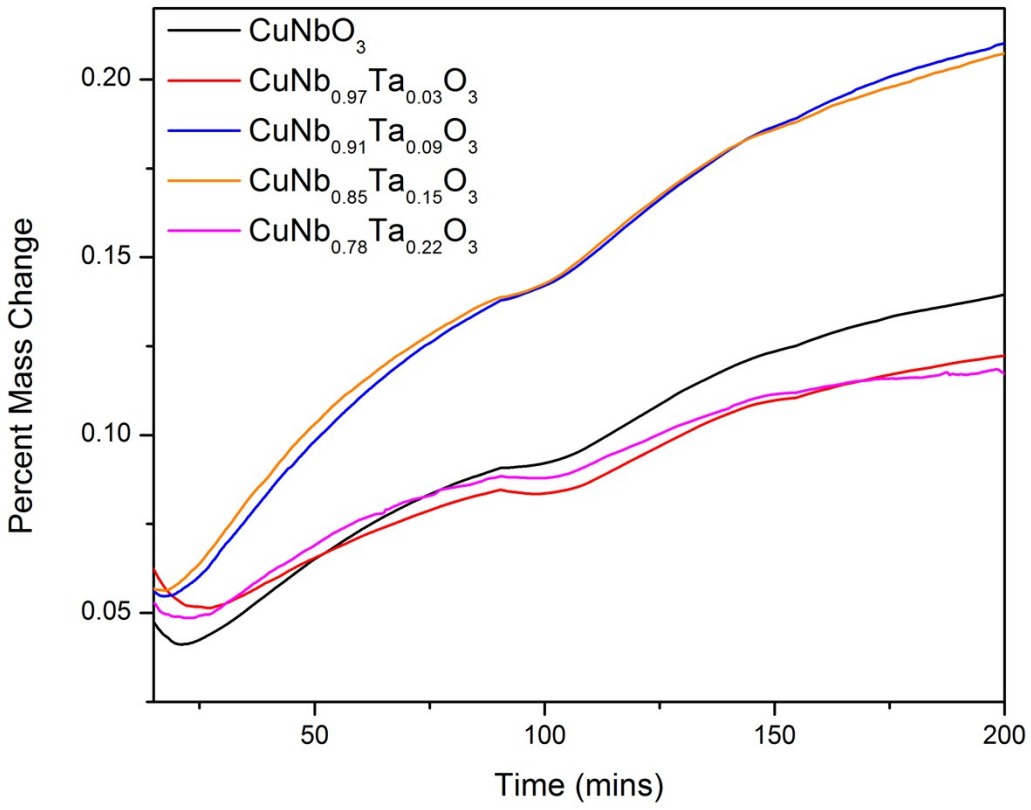


**Figure S3.** Weight gain versus time when heating $CuNb_{1-x}Ta_xO_3$ ($x$ = 0, 0.03, 0.09, 0.15, 0.22) at 250 °C for 3 hrs in air.

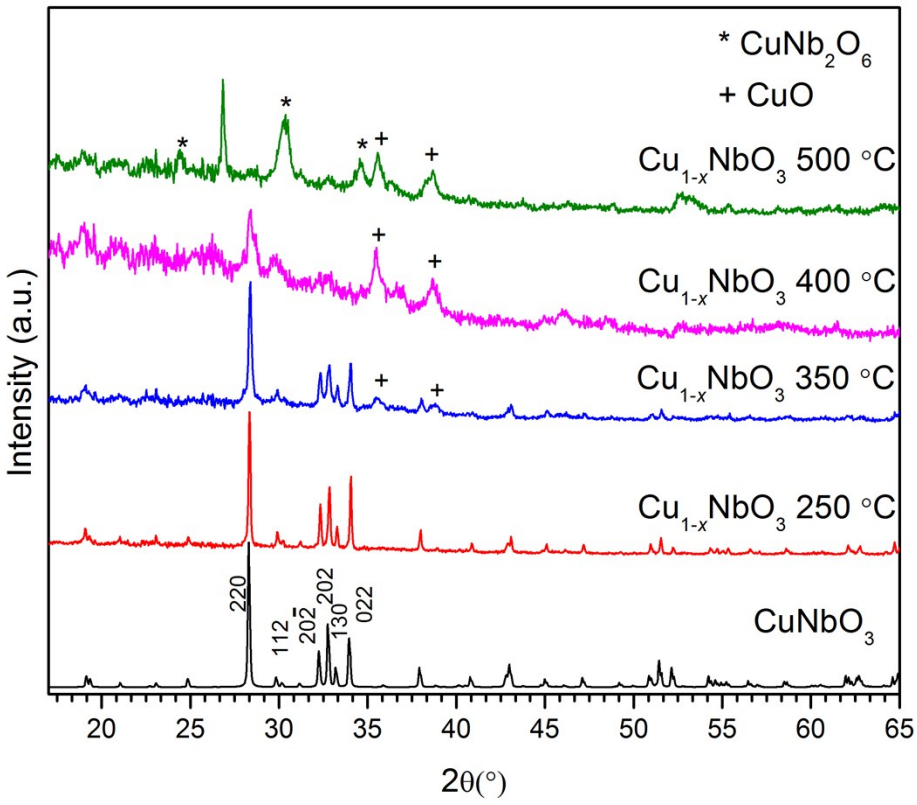


**Figure S4.** Powder X-ray diffraction of $Cu_{1-x}NbO_3$ films heated at 250, 350, 400, and 500 °C for 3 hours. New peaks corresponding to CuO and $CuNb_2O_6$ were identified at temperatures of 350 °C and higher.

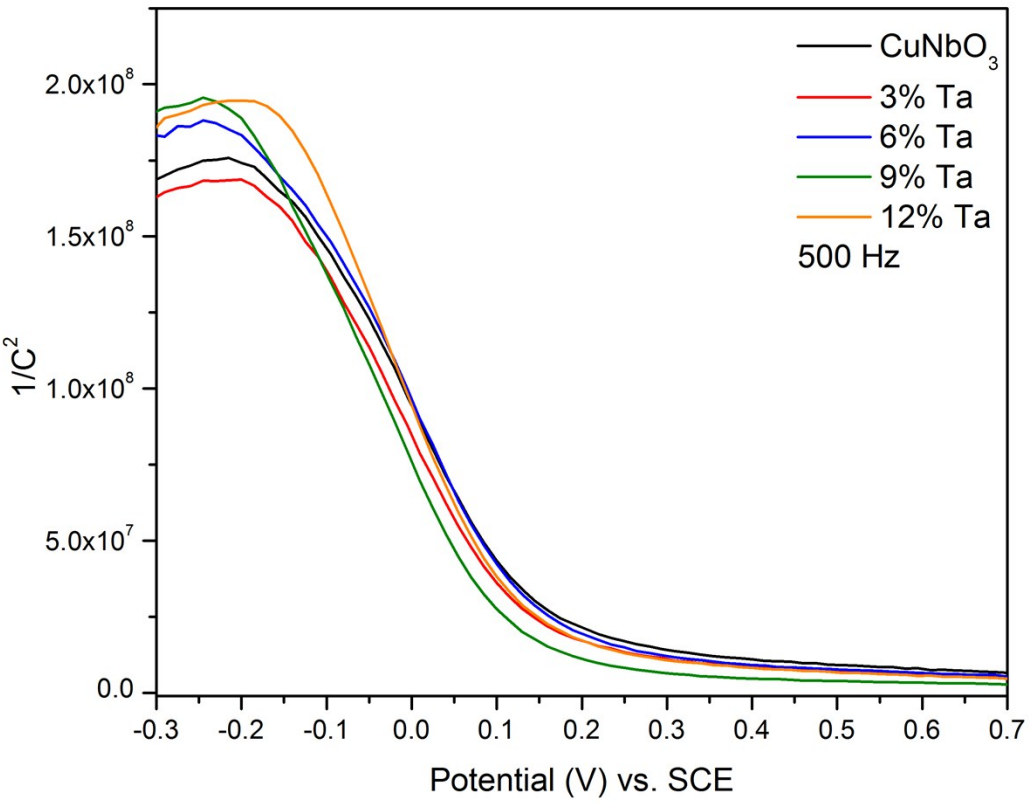


**Figure S5.** Mott-Schottky measurements of $CuNb_{1-x}Ta_xO_3$ ($x$ = 0, 0.03, 0.06, 0.09, and 0.12) with a fixed AC frequency of 500 Hz.

**Mott-Schottky Analysis and Equations**

*Acceptor Density ($N_A$)*

The charge acceptor density ($N_A$) within the metal oxide is calculated from the equation below.

$$N_A = \frac{2}{e\varepsilon_r\varepsilon_0 A^2 m}$$

The slope of the linear potion of each plot (m) is determined and used in the equation. The value of the dielectric for all the samples is approximated to 35 F/m and the area of the films studied was ~4 $cm^2$.

*Energy of the valence band ($E_v$)*

The final calculation for the energy position of the valence band is done with the equation shown below.

$$E_v = E_{fb} + kT \ln(N_A/N_v)$$

In the equation, the effective density of states for the valence band ($N_v$) is approximated to $10^{19}$ $cm^{-3}$.