Supporting Information

**Electrodeposited Thin-Film Cu,Sb Anodes for Li-ion Batteries: Enhancement of Cycle Life via Tuning of Film Composition and Engineering of the Film-Substrate Interface**

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**Fig. S1** Auger depth profiles of amorphous Sb@Cu anodes before (A) and after (B) cycling in a Li-ion half-cell.

**Fig. S2** (A) Differential capacity plot of an amorphous Sb@Ni anode cycled in a Li-ion half-cell. (B) PXRD of the same Sb@Ni anode both before and after cycling.
Fig. S3 (A) Representative EDS spectrum used to quantify the composition of the electrodeposited films, (B) Composition of the amorphous Cu$_x$Sb films as measured by EDS both before and after cycling in Li-ion half-cells. Note that carbon was not included in the quantification of the composition, and that the large oxygen composition is likely due to the presence of a fairly reactive SEI layer on the surface of the thin films.
Fig. S4 Differential capacity plots for the first 50 cycles of the amorphous Cu-Sb@Ni anodes in Li-ion half-cells. The compositions of the anodes are: (A) Sb-only, (B) Cu$_{0.04}$Sb, (C) Cu$_{0.30}$Sb, (D) Cu$_{0.89}$Sb, (E) Cu$_{1.38}$Sb, (F) Cu$_{1.86}$Sb.

Fig. S5 Composition of the crystalline Cu$_x$Sb@Ni films as measured by EDS both before and after cycling in Li-ion half-cells (see explanation in the caption for Fig. S3).
Fig. S6 Auger depth profile of a crystalline Sb@Cu anode before cycling.

Fig. S7 PXRD of crystalline anodes before and after cycling in Li-ion half-cells.
**Fig. S8** Cycle lifetime plot comparing crystalline anodes with the following architectures: Sb@Cu, Sb@Ni, Sb@Ni@Cu.

**Fig. S9** SEM images comparing a pre- (left) and post-cycled (right) Ni-blocking layer.