## **Supplemental document**



FIG. 1. Schematic illustration of CoPi films and Co3O4 film preparation process by ALD



FIG. 2. a., c.): Evolution of cyclic voltammograms with increasing number of CV cycles for the best performing CoPi film, as well as an ALD  $Co_3O_4$  reference. b, d.) Evolution of the OER current density (measured at 1.8 V vs. RHE) as a function of increasing number of CV cycles for CoPi and  $Co_3O_4$ 



Figure 3: Evolution of the Co-to-P and Co-to-O ratios as measured by XPS a function of the number of cyclic voltammetry cycles, compared to the evolution of the OER current density as function of the same.



Figure 4: Relationship between the ECSA and the OER current density for ALD CoPi films with different initial compositions both before and after activation by 500 CV cycles.