## Electronic Supplementary Information for

## Activating Earth-Abundant Electrocatalysts for Efficient, Low-Cost Hydrogen Evolution/Oxidation: Sub-Monolayer Platinum Coatings on Titanium Tungsten Carbide Nanoparticles

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**Fig. S1.** Atomic ratio of surface ML coverage to the total number of metal atoms in the NP versus total NP diameter. Small core-shell NPs with sub-ML NM surface coverages offer complete NM dispersion while also offering bifunctional surfaces with exposed NM sites and TMC sites accessible for catalytic transformations. However, small core-shell NPs with complete ML NM coatings cannot offer substantial NM loading reductions. As such, this regime is designated as "Sub-ML Efficient." For large NPs, complete ML and multilayer NM surface coverages can still result in substantial reductions in NM loadings and are more appropriate for applications in electrocatalysis where durability is a significant challenge. As such, this regime is designated as "ML Efficient."



**Fig. S2.** Transmission electron micrograph of (a) 0.75 ML Pt/TiWC and (b) commercial 20 wt% Pt/C catalysts.



Fig. S3. Pt 4 f signal in the XPS spectrum of 0.05 ML Pt/TiWC.



**Fig. S4.** (a) Schematic representation, (b) STEM image, (c) EDX map, and (d) line scan of a 2 ML Pt/TiWC nanoparticle.



**Fig. S5.** Technoeconomic comparison of earth-abundant CoP catalyst with Pt-containing catalysts expressed as HOR anode lifetime cost vs. Pt loading. Error bars represent prediction intervals with 95% confidence.