Electronic Supplementary Information

Magnetic CoPt Nanoparticle Decorated Ultrathin Co(OH)₂ Nanosheets: An Efficient Bi-functional Water Splitting Catalyst

Bibhudatta Malik,^{\$‡} S. Anantharaj,^{1†‡} K. Karthick, ^{1†} Deepak K. Pattanayak¹, ¹* and Subrata Kundu^{1†#*}

^IAcademy of Scientific and Innovative Research (AcSIR), CSIR-Central Electrochemical Research Institute (CSIR-CECRI) Campus, New Delhi, India

^{\$}Chlor-alkali Division, CSIR-Central Electrochemical Research Institute (CECRI), Karaikudi-630006, Tamil Nadu, India

[†]Electrochemical Materials Science (ECMS) Division, CSIR-Central Electrochemical Research Institute (CECRI), Karaikudi-630006, Tamil Nadu, India

[#]Department of Materials Science and Mechanical Engineering, Texas A&M University, College Station, Texas, TX-77843, USA

[‡]These authors have contributed equally.

* To whom correspondence should be addressed, *E-mail: <u>pattanayak1977@gmail.com</u>* and <u>kundu.subrata@gmail.com</u>, Phone: (+ 91) 4565-241397 and (+ 91) 4565-241487.

Reagents and Instruments

Cobalt(II) acetate hexahydrate, hexachloroplatinic acid, commercial Pt/C 10 wt. % catalyst, potassium hydroxide, 5% Nafion solution and isopropyl alcohol were procured from Sigma-Aldrich. Commercial RuO₂ catalyst was obtained from Alfa Aesar. Polyvinyl pyrrolidone (PVP) was purchased from SRL, India. Hg/HgO reference electrode, Pt-foil counter electrodes were purchased from CH Intruments pvt. Ltd. Carbon cloth (CC) was as working electrode after modifying with the catalysts. Milli pore water (18 M Ω) was used for the entire synthesis and electrocatalysis processes. The synthesized CoPt@Co(OH)2 arrays were characterized with HR-TEM, (TecnaiTM G² TF20) working at an accelerating voltage of 200 kV. The Energy Dispersive X-ray Spectroscopy (EDS) analysis was done with the FE-SEM instrument (Oxford) with a separate EDS detector connected to that instrument. The XRD analysis was done with a scanning rate of 5° min⁻¹ in the 2θ range 10-90° using a Bruker X-ray powder diffractometer (XRD) with Cu K_a radiation ($\lambda = 0.154$ nm). X-ray photoelectron spectroscopic (XPS) analysis was performed using a Theta Probe AR-XPS system (Thermo Fisher Scientific, UK). Electrochemical analyzer CHI6084c version 12.13 was used for the entire electrochemical characterizations. Hg/HgO reference electrode was used along with a Pt-foil counter electrode where our CoPt@Co(OH)₂ arrays modified GC electrode was used as working electrode.

Sample preparation for various characterizations

As synthesized samples were directly taken for XRD and XPS analyses before cycling. TEM samples were prepared by dispersing required quantity in 1 mL of Milli Q water followed by drop casting on carbon coated copper TEM grid and dried at RT for slow evaporation of water before analyses in dark and vacuum. The catalyst ink for OER studies for all four catalysts were prepared by taking 3 mg of catalyst in 1 mL mixture of water, isopropyl alcohol and 5 wt. % Nafion solution in 6.5:2.0:0.5 ratio. The whole content was homogenized in an ultrasonic bath for 30 min and the obtained ink was used for all the electrochemical studies.



Figure S1: Optical images showing magnetic behavior of CoPt@Co(OH)₂ when a magnetic is swept behind the paper.



Figure S2: XRD pattern of CoPt/Co(OH)₂ at 2 min⁻¹.



Figure S3: Type I BET adsorption isotherm of CoPt@Co(OH)₂ nano arrays.



Figure S4: XPS survey scan of CoPt@Co(OH)₂ nano arrays.



Figure S5: High resolution XPS spectrum of C 1s state of C in CoPt@Co(OH)₂ nano arrays.



Figure S6: High resolution XPS spectrum of O 1s state of C in CoPt@Co(OH)₂ nano arrays.



Figure S7: (a-b) The comparative OER polarization and steady state polarization curves of CoPt@Co(OH)₂ and the synthesized Ni(OH)₂.



Figure S8: Optical images showing the bifunctional activity of $CoPt@Co(OH)_2$ arrays equipped with AAA alkaline battery of optimum potential of 1.5 V.



Figure S9: (a-b) Nyquist plots of CoPt@Co(OH)₂ nano arrays modified interface before and after aging of 5 days in 1 M KOH with and without an external magnetic support.