Electronic Supplementary Material (ESI) for Sustainable Energy & Fuels.

Supporting Information

One-Step Synthesis of Graphitic Carbon-Nitride doped with Black-Red Phosphorus as a Novel, Efficient and Free-Metal Bifunctional Catalyst and its Application for Electrochemical Overall Water Splitting

Fatemeh Shiravani, Javad Tashkhourian* and Behzad Haghighi*

Department of Chemistry, Faculty of Sciences, Shiraz University, Shiraz 71454, Iran

* Corresponding authors:
Tel: +98 71 3613 7141, Fax: +98 71 3646 0788
E-mail address: tashkhourian@shirazu.ac.ir (J. Tashkhourian, Orcid ID: 0000-0003-3437-0782)
Tel: +98 71 3613 7178, Fax: +98 71 3646 0788
E-mail address: bhaghighi@shirazu.ac.ir (B. Haghighi, Orcid ID: 0000-0003-0974-7912)

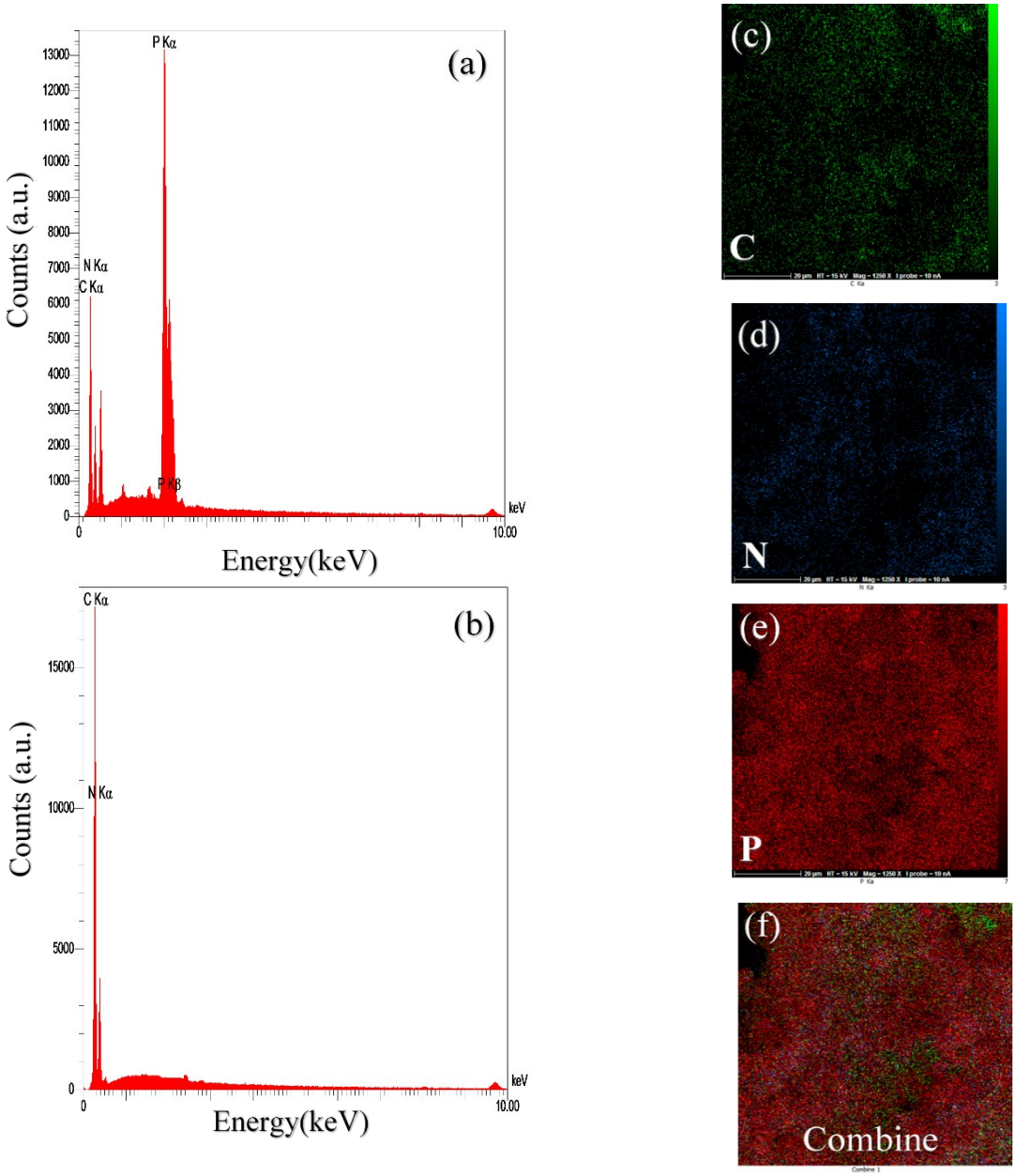

Figure S1 EDS analysis for (a) BRP-g-C_3N_4 and (b) g-C_3N_4, (c-f). Elemental mapping of (c) C, (d) N, (e) P, and (f) C, N and P altogether elements in BRP-g-C_3N_4 composite

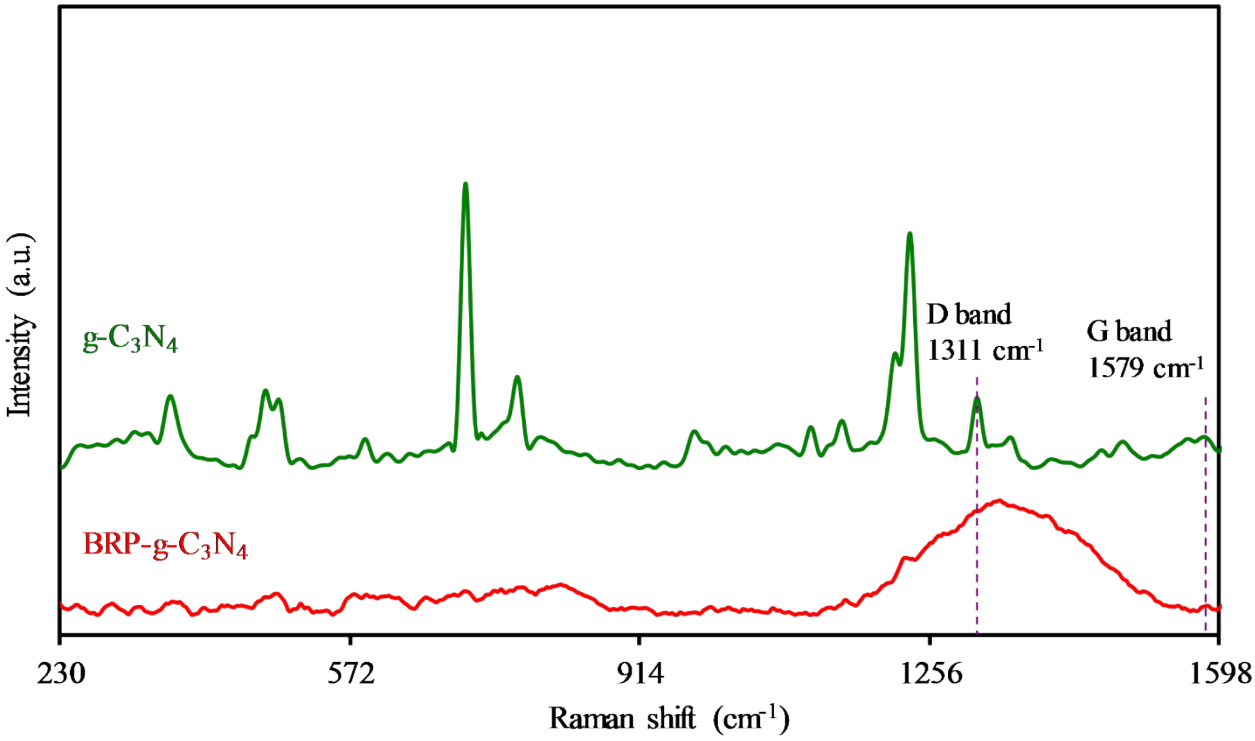

Figure S2 Raman spectra and I_D /I_G ratio for g-C_3N_4, and BRP-g-C_3N_4 composite

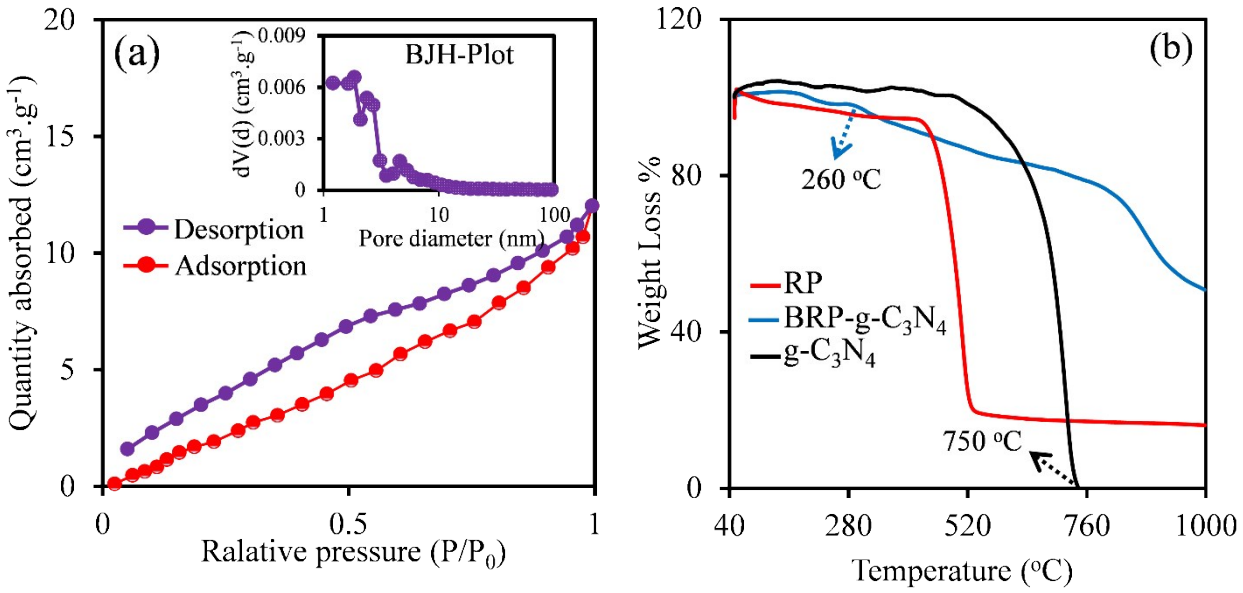

Figure S3 (a) N_2 adsorption−desorption isotherm and (the insert) corresponding pore size distribution of BRP-g-C_3N_4 composite, and (b) TGA curves of RP, BRP-g-C_3N_4, and g-C_3N_4

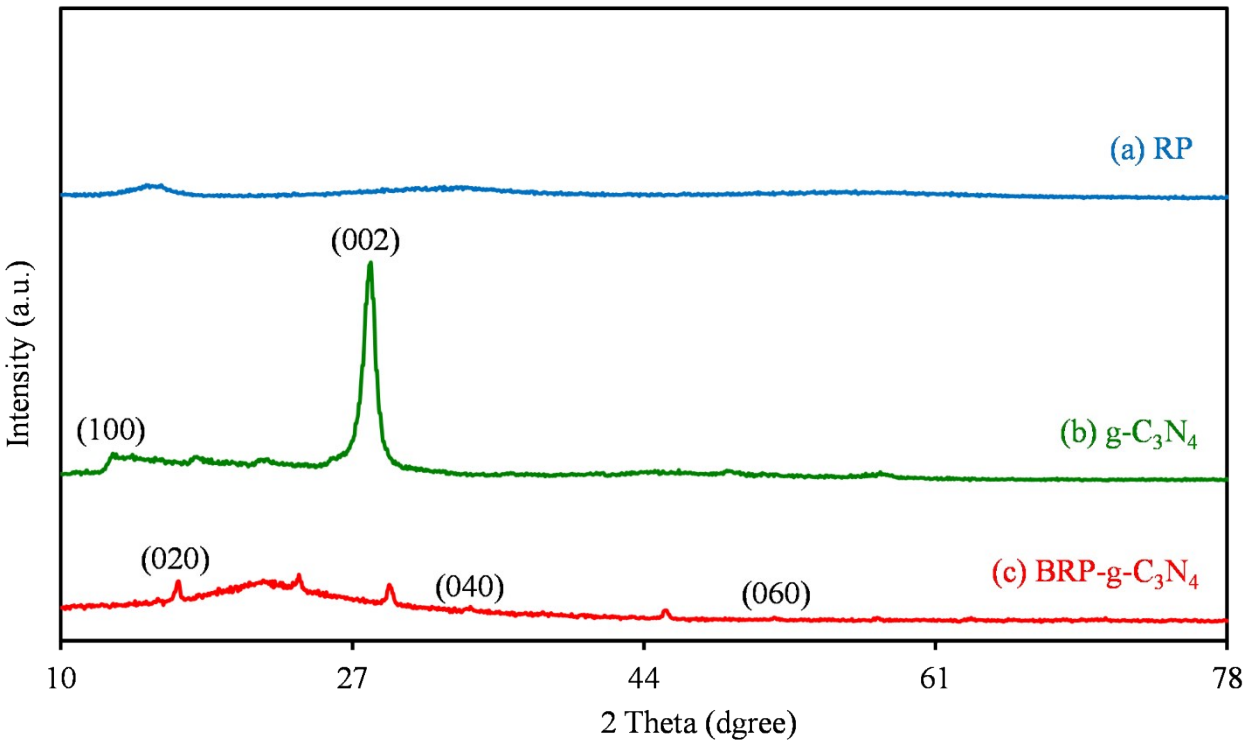

Figure S4 XRD spectra of the (a) RP, (b) g-C_3N_4, and (c) BRP-g-C_3N_4

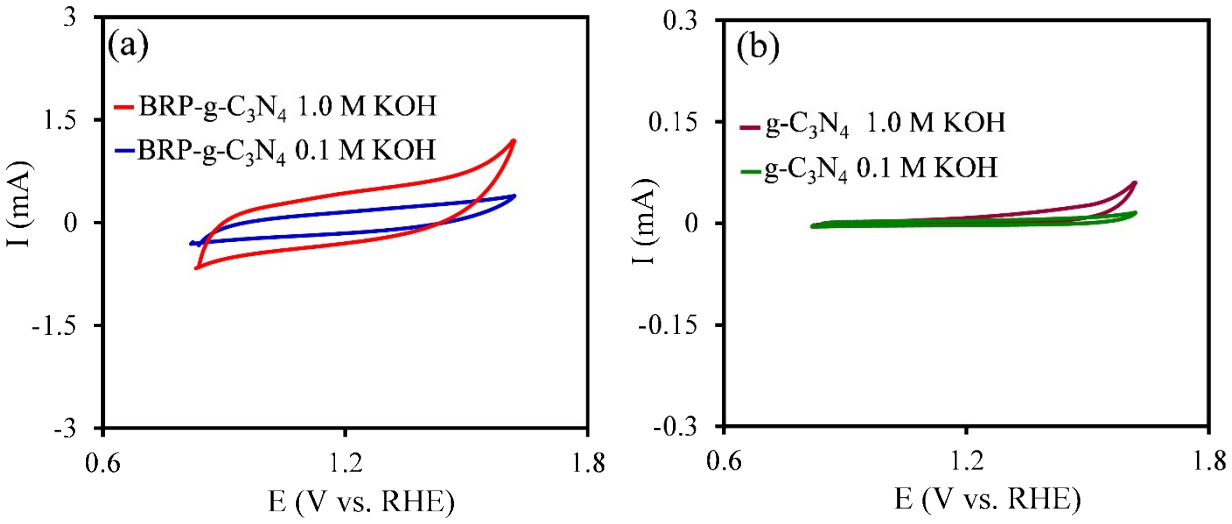

Figure S5 Cyclic voltammograms of (a) BRP-g-C_3N_4, and (b) g-C_3N_4 modified CPE at scan rate 10 mV.s^{-1} in 1.0 and 0.1 M KOH.

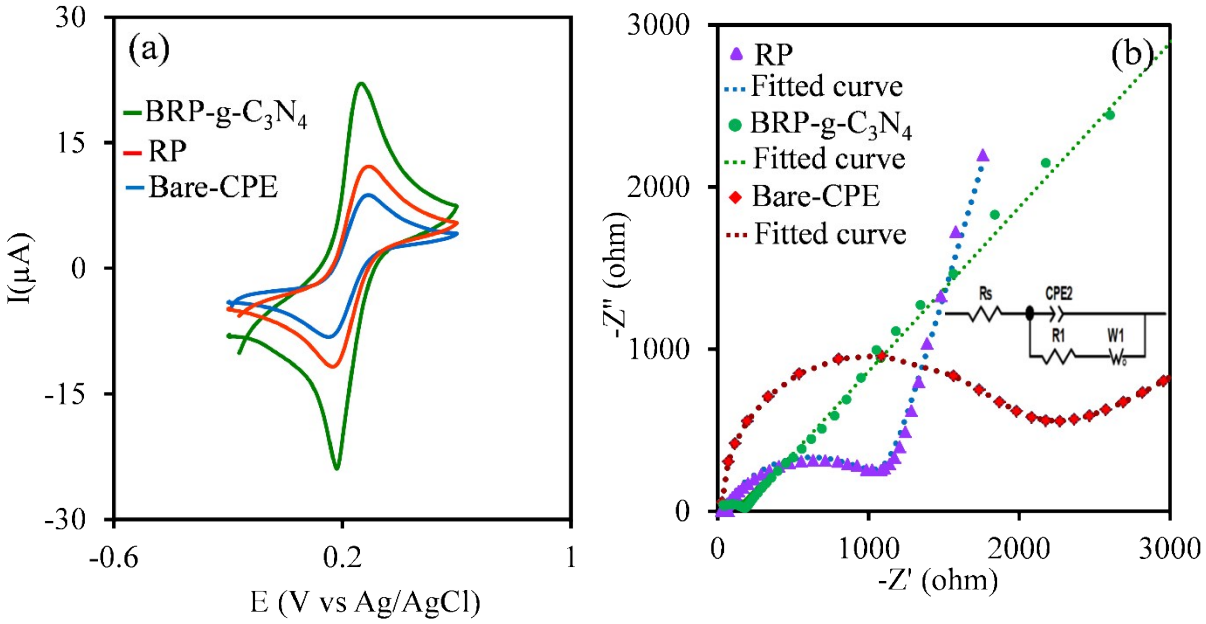

Figure S6 (a) Cyclic voltammograms and (b) electrochemical impedance spectra recorded for BRP-g C_3N_4, RP modified and bare-CPE in 0.1 M KCl containing 5.0 mM $[Fe(CN)_6]^{-3/-4}$. Scan rate for CV studies; 10 mV s^{-1}. Inset: Equivalent circuit

Table S1:

Electrodes	**$R_{ch(ohm)}$**	**$R_{s(ohm)}$**	**CPE-T**	**Chi-Squared**
Bare-CPE	1827	32.39	3.0098E-7	3.8E-6
RP-CPE	1141	29.58	4.1E-5	4.0E-4
BRP-g-C_3N_4-CPE	112	42.6	2.08E-7	1.08E-4

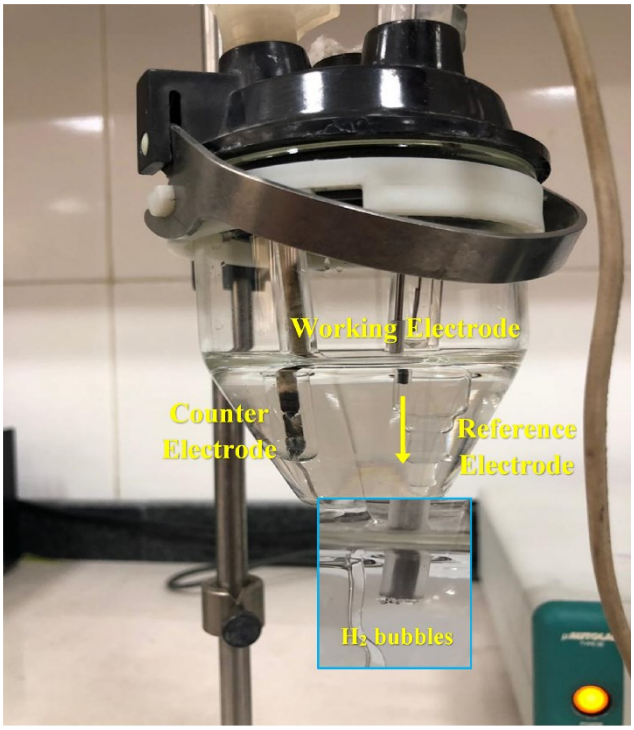

Figure S7 Experimental setup and (the insert) hydrogen bubbles on BRP-g-C_3N_4-CPE.

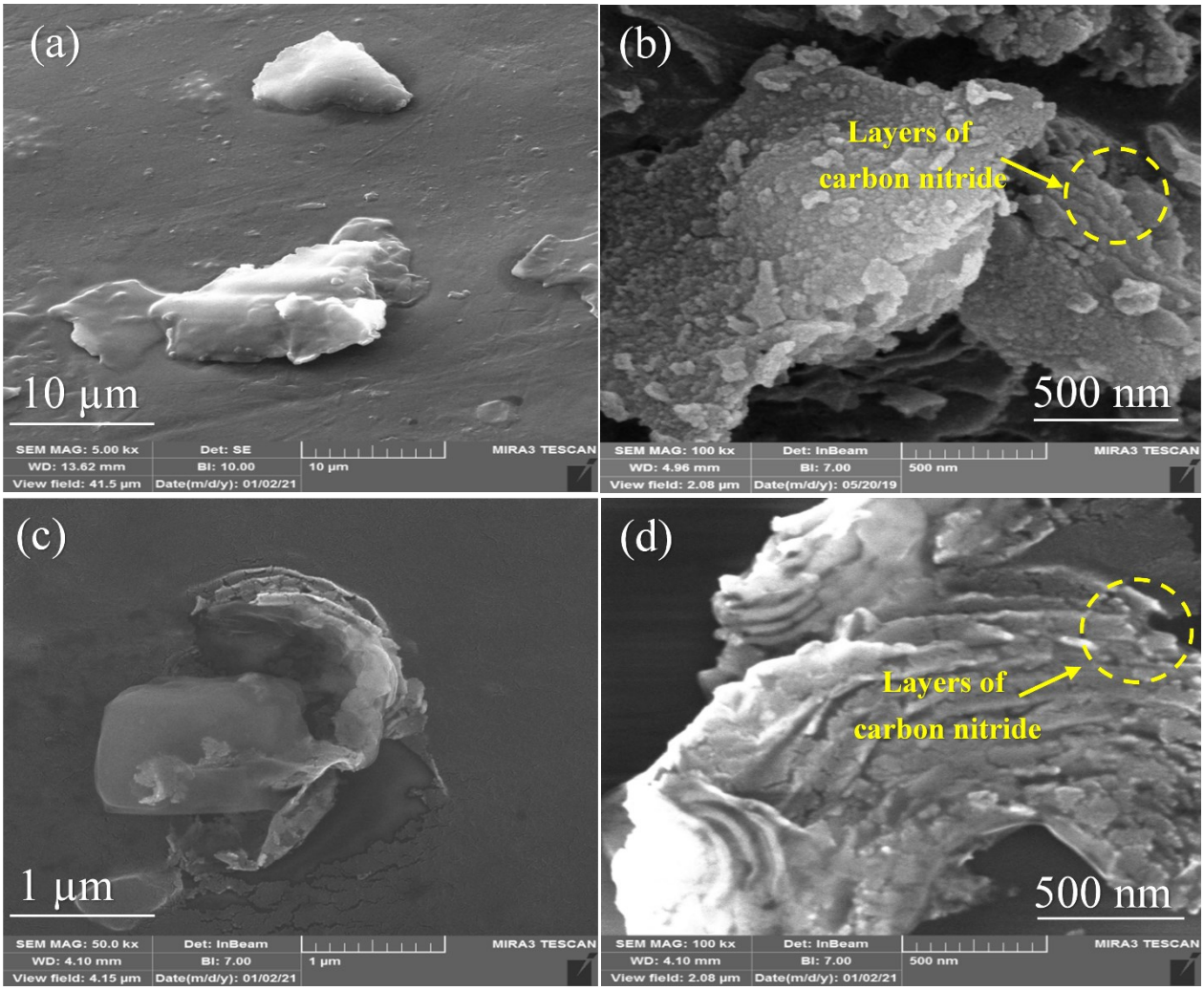

Figure S8 FE-SEM images of BRP-g-C_3N_4-CPE (a,b) before ,and (c,d) after chronoamperometry

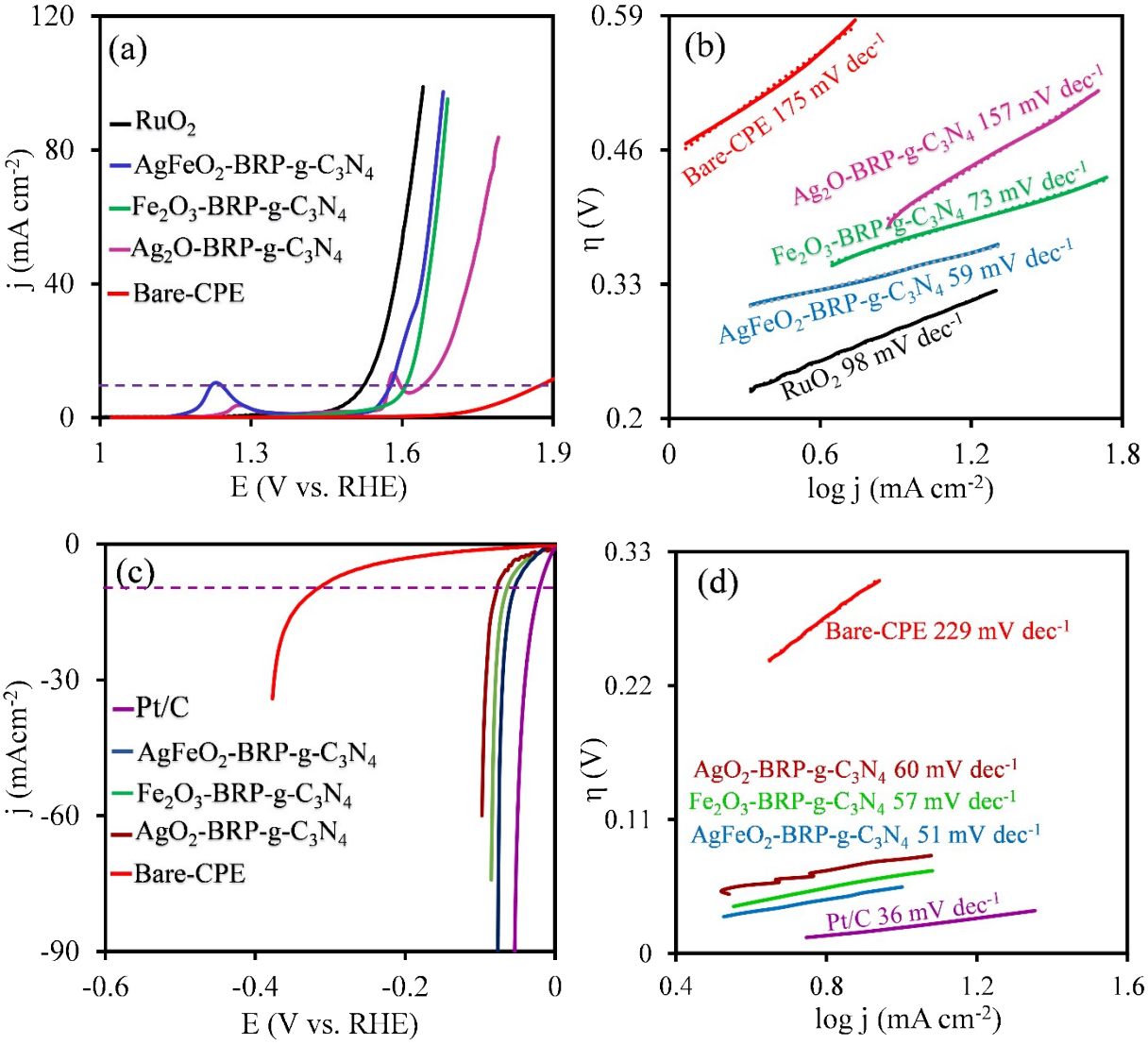

Figure S9 (a) OER polarization curves, (b) corresponding OER Tafel plots, (c) HER polarization curves and (d) corresponding HER Tafel plots for $AgFeO_2$-BRP-g-C_3N_4, Fe_2O_3-BRP-g-C_3N_4, Ag_2O-BRP-g-C_3N modified CPE and bare, RuO_2 and Pt/C CPE in 1.0 M KOH with a scan rate of 5.0 mV s^{-1}.