## Urchin-like hierarchical ruthenium cobalt oxide nanosheets on $Ti_3C_2T_x$ MXene as

## binder-free bifunctional electrode for overall water splitting and supercapacitors

Parvin Asen<sup>1</sup> and Ali Esfandiar<sup>1,\*</sup>, H. Mehdipour<sup>2,3</sup>

<sup>1</sup>Department of Physics, Sharif University of Technology, Azadi Street, 11365-9161, Tehran, Iran

<sup>2</sup>Faculty of Physics, University of Duisburg-Essen, Lotharstr. 1, 47057 Duisburg, Germany

<sup>3</sup>Department of Chemistry, Amirkabir University of Technology, Tehran, Iran

Email: esfandiar@physics.sharif.edu

Fig. S1: Chronoamperometric data for electrodeposition of  $RuCo_2O_4$  on  $Ti_3C_2T_x$  MXene@NF electrode

Fig.S2: DFT simulation of RuCo<sub>2</sub>O<sub>4</sub>/Ti<sub>3</sub>C<sub>2</sub> interface

Fig.S3: DFT-computed relative total energy of RuCo<sub>2</sub>O<sub>4</sub>

**Fig.S4:** EDS data of  $Ti_3C_2T_x$  on NF electrode

Fig.S5: SEM, elemental mapping images and EDS of electrodeposited  $RuCo_2O_4$  on  $Ti_3C_2T_x@NF$  at 100 s

Fig.S6: SEM, elemental mapping images and EDS of electrodeposited  $RuCo_2O_4$  on  $Ti_3C_2T_x$  @NF at 500 s

Fig.S7: SEM and EDS analysis of  $RuCo_2O_4@NF$  and SEM images of  $Co_3O_4/Ti_3C_2T_x$  MXene@NF,  $RuO_2/MXene@NF$ ,  $Co_3O_4@NF$ ,  $RuO_2@NF$ 

**Fig.S8:** LSV curves of RuCo<sub>2</sub>O<sub>4</sub>/Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> MXene@NF electrode towards OER and HER at various deposition time of RuCo<sub>2</sub>O<sub>4</sub>

**Fig.S9:** CV curves of  $Ti_3C_2T_x$  MXene@NF, RuO<sub>2</sub>@NF, RuO<sub>2</sub>/ $Ti_3C_2T_x$  MXene@NF, RuCo<sub>2</sub>O<sub>4</sub>@NF, Co<sub>3</sub>O<sub>4</sub>@NF, Co<sub>3</sub>O<sub>4</sub>/ $Ti_3C_2T_x$  MXene@NF, and RuCo<sub>2</sub>O<sub>4</sub>/ $Ti_3C_2T_x$  MXene@NF at scan rate of 20 mV s<sup>-1</sup>

Fig.S10: CV curves of RuCo<sub>2</sub>O<sub>4</sub>/Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> MXene@NF at various deposition time of RuCo<sub>2</sub>O<sub>4</sub>

Table S1: Fitting data of the Nyquist plots



Fig.S1 I-t curve for electrodeposition of  $RuCo_2O_4$  on  $Ti_3C_2T_x$  MXene@NF



**Fig. S2** DFT model of RuCo<sub>2</sub>O<sub>4</sub>/Ti<sub>3</sub>C<sub>2</sub> interface: plane (a) and side (b) views of relaxed geometries of two-molecule RuCo<sub>2</sub>O<sub>4</sub> cluster on a Ti<sub>3</sub>C<sub>2</sub> (001) surface. Light blue, green, gray, dark blue and red balls represent Ti, C, Co, Ru and O atoms, respectively



**Fig. S3** Computed relative total energy of RuCo<sub>2</sub>O<sub>4</sub> structure as a function of cut-off energy of expansion of electron wave function. The reference energy is the total energy of the structure for cut-off of 100 Ry.



Fig.S4 EDS analysis of Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> MXene@NF



Fig.S5 (a) SEM image of  $RuCo_2O_4/Ti_3C_2T_x$  MXene@NF at electrodeposition time of 100 s of  $RuCo_2O_4$  on  $Ti_3C_2T_x$  MXene@NF, and (b-h) SEM images and EDS analysis of the fabricated electrode



**Fig.S6** (a, b) SEM image of  $RuCo_2O_4/Ti_3C_2T_x$  MXene @NF at electrodeposition time of 500 s of  $RuCo_2O_4$  on  $Ti_3C_2T_x$  MXene@NF, and (c-i) SEM images and EDS analysis of the fabricated electrode



**Fig.S7** SEM images of (a) Co<sub>3</sub>O<sub>4</sub>/Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> MXene@NF, (b,c) RuO<sub>2</sub>/Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> MXene@NF, (d ,e) SEM image and EDS analysis of RuCo<sub>2</sub>O<sub>4</sub>@NF, (f, g) Co<sub>3</sub>O<sub>4</sub>@NF and (h) RuO<sub>2</sub>@NF



**Fig.S8** (a) OER and (b) HER curves of  $RuCo_2O_4/Ti_3C_2T_x$  MXene @NF at different electrodeposition times of  $RuCo_2O_4$  (100, 300 and 500 s) on the  $Ti_3C_2T_x$  MXene@NF electrode



 $\label{eq:Fig.S9} \begin{array}{l} Fig.S9 \ CV \ curves \ of (a) \ Ti_3C_2T_x \ MXene@NF, \ RuO_2@NF, \ RuO_2/ \ Ti_3C_2T_x \ MXene@NF, \ (b) \\ RuCo_2O_4@NF, \ Co_3O_4@NF, \ Co_3O_4/ \ Ti_3C_2T_x \ MXene@NF, \ (c) \ RuCo_2O_4/ \ Ti_3C_2T_x \\ MXene@NF, \ Co_3O_4/ \ Ti_3C_2T_x \ MXene@NF \end{array}$ 



Fig.S10 CV curves of RuCo<sub>2</sub>O<sub>4</sub>/Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> MXene@NF at different electrodeposition times of RuCo<sub>2</sub>O<sub>4</sub> (100, 300 and 500 s) on the Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> MXene@NF electrode surface at scan rate of  $20 \text{ mV s}^{-1}$ 

## Table S1 Fitting parameters used to simulate the EIS data obtained for prepared electrodes in alkaline media

Samples	R <sub>s</sub> (Ω)	R <sub>1</sub> (Ω)	CPE1 (Ω <sup>-1</sup> s)	n1	W	n	CPE2 (Ω <sup>-1</sup> s)	n2
RuCo <sub>2</sub> O <sub>4</sub> / Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> MXene@NF	0.005	1.2	2	0.95	0.06	0.4	1	0.4
RuO <sub>2</sub> / Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> MXene@NF	0.67	2	0.18	0.65	0.06	1	1.3	0.8
C03O4/ Ti3C2Tx MXene@NF	0.005	4	0.6	0.8	0.02	0.39	0.4	0.7
RuCo <sub>2</sub> O <sub>4</sub> @NF	0.5	4	0.62	0.81	0.02	0.5	0.15	0.6
Co <sub>3</sub> O <sub>4</sub> @NF	1	8	0.61	0.83	0.02	0.35	0.6	0.8
RuO <sub>2</sub> @NF	1	45	0.05	0.8	0.009	0.34	7	0.4
Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> MXene@NF	1	75	0.03	0.8	0.002	0.3	15	0.4