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Supporting information

Supercapacitors with Enhanced Energy Storage and Hydrogen Evolution Reaction Performance via Sequential Alkali-Modified MXenes

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1. MXene synthesis

 $Ti_3C_2T_x$ MXene was synthesized by etching the Ti_3AlC_2 MAX phase (Carbon Ukraine Ltd) using an HCl/LiF etchant. LiF powder (1.6g) was mixed with 30 ml HCl (12M) for 10 min, then Ti_3AlC_2 (1.6g) was added and stirred for 48h at 40°C. The mixture was centrifuged at 3500 rpm and washed with DI water until pH ~6. The greenish-black MXene mixture was mildly sonicated in DI water for 30 min, then centrifuged at 2000 rpm for 30 min. The resulting MXene was termed as MX-1.

2. Characterizations

Structural and morphological characteristics of all samples were analyzed using a Hitachi SU8010 field emission scanning electron microscope (FESEM) and their elemental compositions were determined through energy-dispersive X-ray spectroscopy (EDX). The crystalline structure of the MXene nanogrooves was examined with a PANalytical X'pert PRO X-ray diffraction (XRD) setup. Raman microscopy was conducted using the Renishaw InVia instrument (UK), to assess the vibrational modes within the samples. The specific surface areas (SSA) of both materials were calculated via Brunauer-Emmett-Teller (BET) analysis plots using a BELSORP-max instrument (BEL Japan Inc.). Electrochemical properties of the symmetric supercapacitors were evaluated through cyclic voltammetry (CV) and galvanostatic charge-discharge (GCD) studies were carried out using (Biologic SP-50) and electrochemical impedance spectroscopy (Bio-logic SP-200)

system in the USA. Interatomic bonds and electronic states of the materials were probed using X-ray photoelectron spectroscopy (XPS), performed with a PHI 5000 Versa Probe III instrument by ULVAC PHI, Japan. Electrochemical measurements (HER) were performed with an electrochemical workstation (Gamry Interface, 1010 E CHI 760E) using a three-electrode system in an aqueous 1.0 M KOH solution.

3. FESEM/TEM and SAED images



Figure S1. (a) FESM images of the MAX phase. TEM images of (b) MX-1 and (c) MX-1-KOH. HRTEM image of (d) MX-1 and (e) MX-1-KOH. SAED image of (f) MX-1 and (g) MX-1-KOH.

4. XRD plots



Figure S2. XRD spectra of Ti_3AlC_2 MAX phase, KOH treated MAX phase i.e. MAX-KOH and MX-1 MXene.

5. XPS plots



Figure S3. XPS Survey spectra for (a) MX-1 (b) MX-MX-1-KOH (c) MX-2 and (d) MX-2-KOH. High-resolution O 1s XPS Survey spectra for (e) MX-1 (f) MX-MX-1-KOH (g) MX-2 and (h) MX-2-KOH.

6. CV cyclic stability curves



Figure S4. Cyclic stability plots for (a) MX-1 (b) MX-MX-1-KOH (c) MX-2 and (d) MX-2-KOH MXene samples at the first and 5000th CV cycle.



Figure S5. (a) Correlation between peak currents and scan rates in CV curves for ion-diffusion estimation. (b) CV, (c) CD, and (d) EIS plots after cyclic stability studies for MX-2-KOH MXene.



Figure S6. (a) Bode plots at different potentials and (b) Turnover frequency (TOF) vs overpotential plot.