Electronic Supplementary Material (ESI) for RSC Advances.

Supporting Information

Reduced graphene oxide/MXene hybrid decorated graphite felt as an effective electrode for vanadium redox flow battery

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1. Model Description

A 3D model of the electrode was established based on the reconstructed electrode pore model. In the applications, the reaction generally occurs on the surface of solid phase. To reveal the physical-chemical process during the electrolysis of bicarbonate, the model was developed based on following assumptions:

(1) The system was isothermal and at steady state.

(2) Ion transport in the electrode was dominated by both diffusion and convection simultaneously.

(3) Electrolyte were assumed as an ideal gas and incompressible.

Mass transport of dissolved species

The dissolved species in electrolyte were solved by using Nernst-Planck equation [28, 29]:

$$0 = -\nabla N_i + S_i \tag{1}$$

$$N_i = -D_i \nabla c_i + u c_i \tag{2}$$

where N_i and R_i are molar flux and reaction source terms, and D_i and u are the diffusion coefficient and electrolyte velocity. The first term on the right side of eq. (1) is the diffusion resulting from concentration difference and the second term the convection from the flow of electrolyte.

The effective mobility $u_{i,e}$ and diffusion coefficient D_i are correlated using Nernst-Einstein equation [30]:

$$u_{i,e} = \frac{D_i}{RT} \tag{3}$$

where R and T are gas constant (8.314 J/(mol·K)) and temperature (K).

The source terms S_i in electrode is calculated based on the current density of electrode:

$$S_i = -\frac{e|i_c|}{n_i F} \tag{4}$$

where e and n_i are the stoichiometric coefficient and numbers of electrons in electrochemical reaction.

Fluid flow

Fluid dynamics in electrode was simulated using laminar flow model, the mass conservation for the electrolyte and gas phases is expressed as:

$$\frac{\partial \rho_l}{\partial t} + \nabla \cdot (\rho_l u_l) = 0 \tag{5}$$

Momentum conservation in the flow field was described using Navier-Stokes equation:

$$\rho(u \cdot \nabla)u = \nabla \cdot [-p + \mu(\nabla u + (\nabla u)^T)] \tag{6}$$

Boundary condition

For the mass transport of chemical species, the flux of species at the wall was set to zero, the bottom of electrode (inflow) was set as a constant concentration boundary, and the outflow of electrolyte was assumed as zero flux:

$$-D_{i,wall}\nabla c_i = 0 \tag{7}$$

$$C_{i,inflow} = C_{i,bulk} \tag{8}$$

$$-D_i \nabla c_i|_{outflow} = 0 \tag{9}$$

To solve the fluid flow equations, the outlet of electrolyte was standard pressure, and the inlet of electrode was set as a constant velocity boundary:

$$p|_{outlet} = p_0 \tag{10}$$

$$u_l|_{inlet} = u_0 \tag{11}$$

Above governing equations and related boundary conditions are solved with COMSOL Multiphysics 6.0 at a steady condition. The parameters of model were summarized and listed in **Table S1**.

Table S1 Model parameters for simulation

Parameter	Value	Unit
Temperature	295.15	K
Pressure	1	atm
Diffusion coefficient of V^{2+}	2.4×10^{-10}	m^2/s
Diffusion coefficient of V^{3+}	2.4×10^{-10}	m^2/s
Electrolyte dynamic viscosity	4.928×10^{-3}	Pa·s
Density of electrolyte	1350	kg/m^3
Initial V^{2+} concentration	257	mol/m^3
Initial V^{3+} concentration	1443	mol/m^3
Inlet velocity	1.42	cm/s

2. Physicochemical characterizations

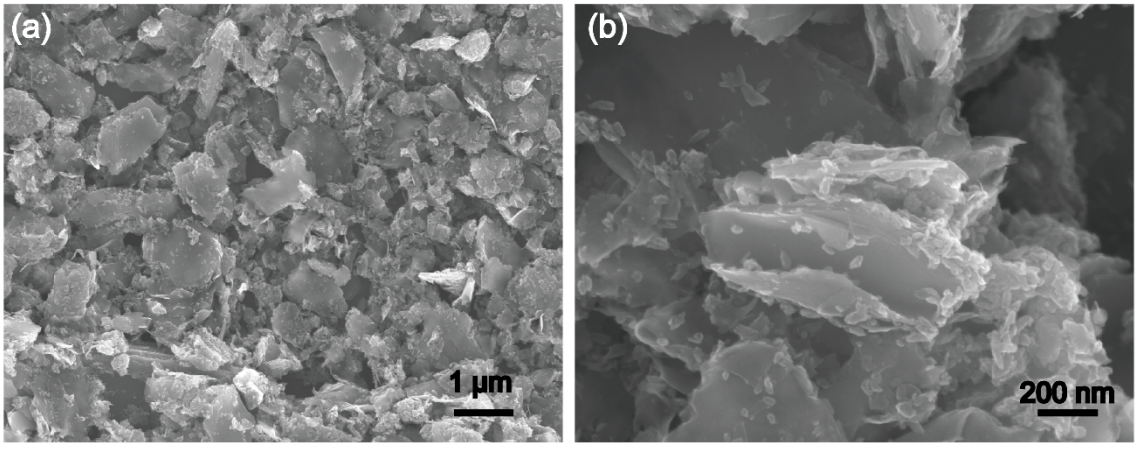

Fig. S1 SEM picture of Mxene at different magnifications

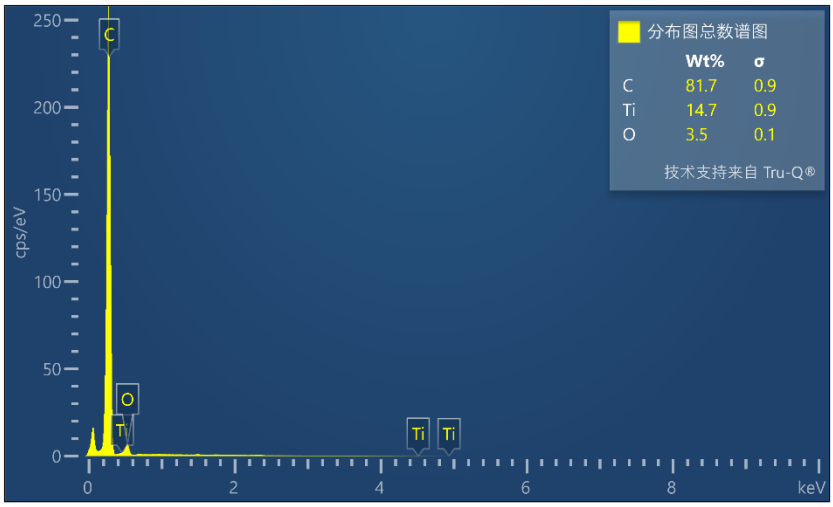

Fig. S2 EDS spectrum of rGO/Mxene@GF

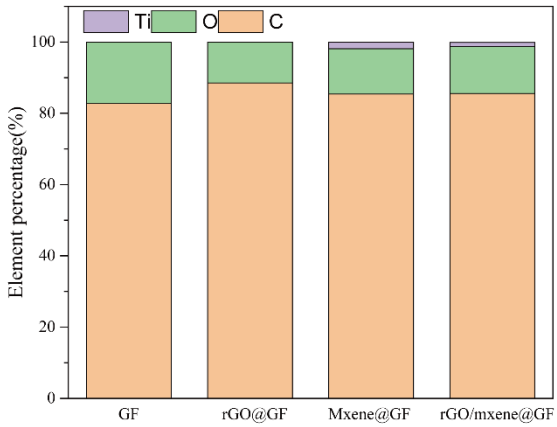

Fig. S3 Elemental composition of electrodes according to XPS measurement.

3. Physicochemical tests

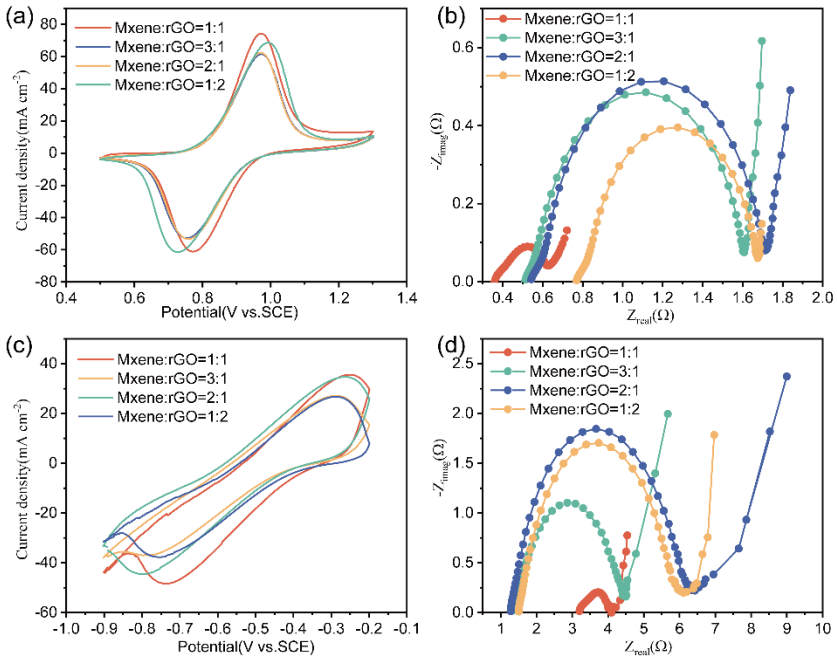

Fig. S4 (a) CV curves of VO^{2+}/VO_2^+ and (b) EIS measurements in 0.1 M VO^{2+} + 3 M H_2SO_4, (c) CV curves V^{2+}/V^{3+} redox reaction and (b) EIS spectrum measurements in 0.1 M V^{3+} + 3.0 M H_2SO_4.

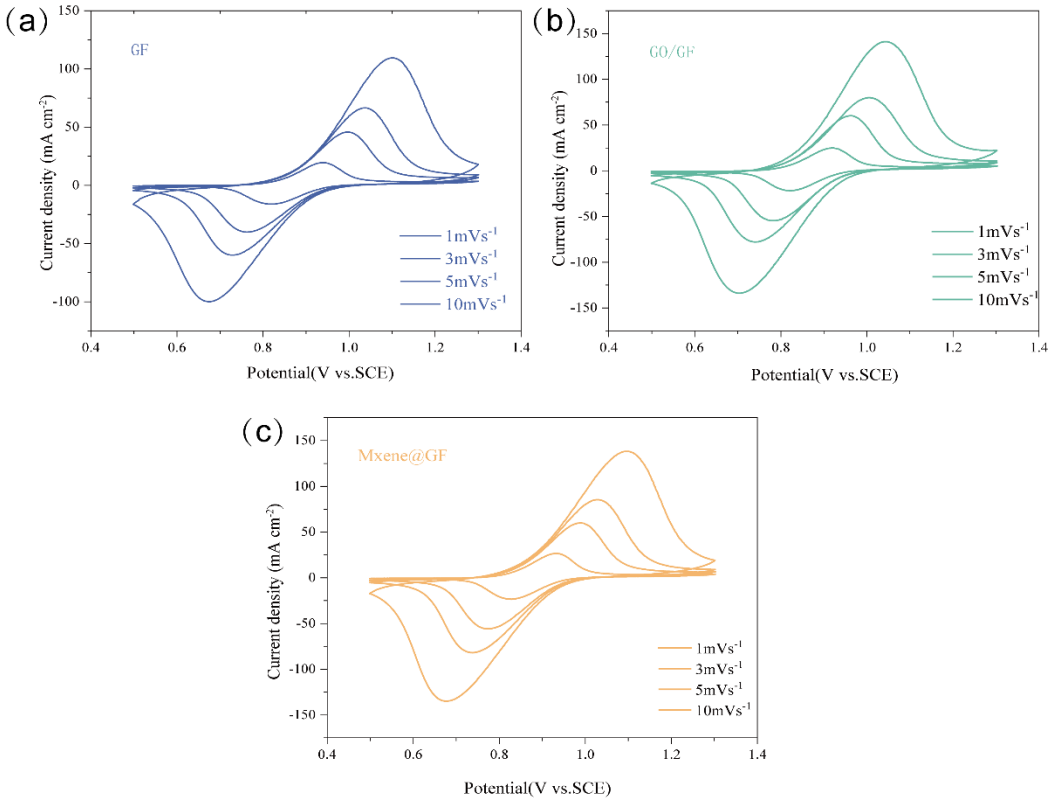

Fig. S5 CV curves of GF, rGO@GF and Mxene@GF at different scan rates.

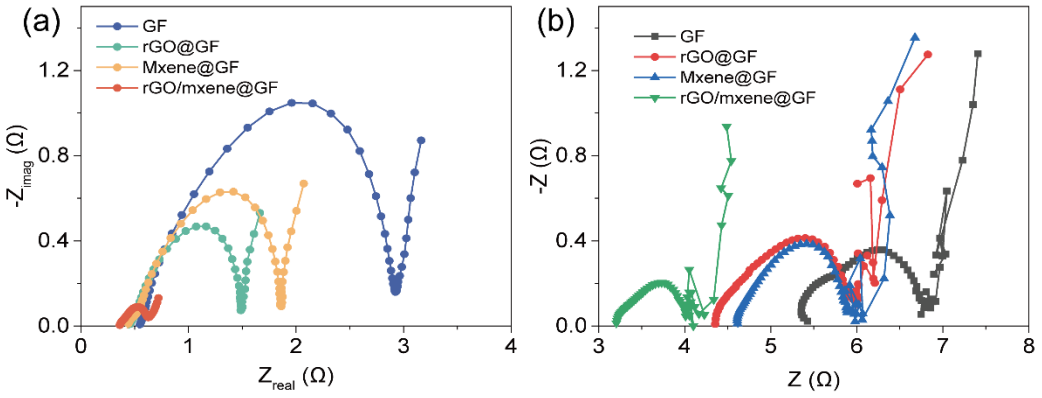

Fig. S6 Nyquist plot of positive and negative electrodes.